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RESEARCH TO INVESTIGATE THE AGING CHARACTERISTICS OF SAMARIUM COBALT MAGNETS

Herbert F. Mildrum, et al

Dayton University

Prepared for:

Air Force Materials Laboratory

March 1974

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RESEARCH TO INVESTIGATE THE AGING CHARACTERISTICS OF SAMARIUM COBALT MAGNETS

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The principal objective of this recently concluded program was to investigate the aging stability of commercially produced sintered $SmCo_5$-based permanent magnets from two domestic sources. The results of measurements of the long-term and elevated-temperature stability of certain engineering design parameters conducted during the program are summarized and reported. Investigations were conducted on cylindrical magnet samples having length-to-

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SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered) diameter ratios corresponding to $B_d/H_d \approx 1/8$, 1/4, 1 and 2-1/2. These investigations included "natural" air and vacuum aging at room temperature; air aging at room temperature after a prestabilizing heat treatment ("thermal knockdown"), and accelerated thermal aging between 150°C and 300°C with and without prior thermal knockdown. Also included were compressive loading strength measurements from room temperature to 250°C. Studies of the thermal aging at temperatures between 150° and 250°C under simultaneous compressive loading of 10 kpsi (700 kg/cm^2) were made. Some samples were exposed to alternating demagnetizing fields varying between approximately $-_BH_c$ and zero (nearly "full recoil"). Effects of material removal by surface grinding or spark discharge were also investigated. Reversible temperature coefficients and irreversible losses were measured during repeated thermal cycling between room temperature, 300°C and -100°C.

SUMMARY

The objective of the work under this contract was the characterization of commercially produced, sintered $SmCo_5$ magnets for various engineering applications. Of particular interest was the long-term stability of selected magnetic properties under environmental influences which simulate the operating conditions of the magnets in devices such as microwave tubes and rotating electrical machines.

The research investigations into the aging characteristics of samarium-cobalt magnets were formulated to provide objective information on the sensitivity of the magnetic properties to:

1) Variables in the basic magnet manufacturing processes
2) Storage under varying conditions
3) Processing of the magnets into usable configurations
4) Usage under various adverse thermal, mechanical and demagnetizing loads representative of the environmental conditions likely to be encountered by the magnets in device applications.

Magnet samples of two principal suppliers were tested: from the General Electric Company and the Raytheon Company. These magnets were produced by somewhat different processes, one involving the isostatic compaction of a two-powder blend (sample code letter A), the other die-pressing of a single-alloy powder (code letter B). Our measurements revealed significant differences between the two sample types with respect to their initial properties and the consistency of these properties in production at the time our samples were made (summer and autumn of 1972). In most cases, however, no clear correlation with sample origin and production procedure could be found regarding the losses on heating, the long-term stability at elevated temperature, or the response to a.c. field cycling. The test sample geometry was axially magnetized cylinders of 1/4 and 1/2 inch diameter of several short axial lengths (thickness).

Environmental exposure tests of the following kinds were conducted. Samples were thermally cycled from room temperature to +300°C and to -100°C, irreversible and reversible losses as well as temperature coefficients were determined. Magnets were aged at temperatures of 25° C, 150°, 200°, 250° and 300°C in an open-circuit condition, with operating points corresponding to unit permeance values of $B_d/H_d \approx 1$, 1/4 and 1/8, and the time dependence of the open-circuit remanent flux was measured. This was done on the as-received samples, on magnets which had previously undergone a thermal stabilization treatment, and on samples which were simultaneously subjected to compressive stresses. In preparation for the latter test, measurements of the mechanical fracture strength under uniaxial compression were made at room and elevated temperatures. Other magnets were subjected to several millions of cycles of an alternating magnetic field to simulate operating conditions in a motor or similar device in which the magnet recoils from close to $_BH_c$ to near H = 0. (Originally planned stability tests under the combined influence of elevated temperature, mechanical pressure and alternating magnetic fields - in a rotating fixture - were abandoned because of the difficulties of meaningfully simulating, or even defining, the operating loads a magnet would experience in a hypothetical large motor or generator.) The effects of machining magnet samples to smaller size - by centerless grinding, flat surface grinding and electric discharge machining - on their elevated-temperature stability were also investigated.

Most measuring procedures and instruments used were described in the first semi-annual progress report. The results of initial-property measurements and their statistical interpretation, thermal cycling data, and information on the aging behavior gained during the first year, were contained in the second report. This information is not repeated here. This third and last report contains data for all the other tests mentioned and the final results of aging investigations which continued to the end of the contract period, even if preliminary data were included in the second report. This final report is thus not necessarily a summary of all significant results obtained under the contract. The previous reports should be consulted for the results of some of the tests, and for experimental details of others whose final outcome is reported in the following pages.

It must be emphasized that the environmental exposure tests were not done on sufficiently large numbers of samples to make them statistically meaningful. While the results clearly point out certain common properties and problems of sintered $SmCo_5$ magnets, the detailed behavior often varied quite drastically within the small group of samples selected for a particular measurement. It would therefore be wrong to say that the set of properties reported for any given sample is characteristic of $SmCo_5$ magnets in general, or even for those made by one of the two principal manufacturing processes.

It is also important to point out that the samples tested are not necessarily representative of the magnets produced by our suppliers at the time this report is issued, 18 to 20 months after our samples were purchased. The manufacturers point out that several changes in their processes have taken place which are said to have improved the performance characteristics and the consistency of their products. Such changes - and progress toward better quality - must, of course, be expected in any new and rapidly developing technology like that of rare earth-cobalt magnets. Proof of the claims made would require measurements and tests on magnet samples from the present production.

FOREWORD

The research investigation described in this report was performed in the Magnetics Laboratory of the University of Dayton by personnel of the Research Institute and the Electrical Engineering Dept. It was sponsored and administered under Contract No. F33615-72-C-1795, Program Code No. FY1457, Project No. 7367, Task No. 736703 by the Air Force Materials Laboratory, Air Force Systems Command, Wright-Patterson Air Force Base, Ohio. Project Engineer: Mr. Donald Evans, AFML/LPE, Tel. (513) 255-4474.

The following personnel participated actively in the investigation during the reporting period: M. S. Hartings, H. F. Mildrum, K.J. Strnat and K. M. Wong.

This report covers research conducted between 1 July 1973 and 30 December 1973. The manuscript was released by the authors in February 1974 for publication as a technical report.

Charles H. Robison

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Chief, Solid State Materials Branch
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SECTION I

INTRODUCTION

The general purpose of the work under this contract was to investigate the stability of commercially produced, sintered, $SmCo_5$-based permanent magnets under various adverse environmental influences. Tests were performed which simulate certain conditions to which magnets are exposed in engineering applications, especially in traveling wave tubes and rotating electrical machines. The contract stipulated as the most important objective the generation of data on the magnetic characteristics and on the aging behavior which is useful to the design engineer. Efforts aimed primarily at an understanding of the physical causes of the deterioration of properties during aging, or at the development of possible remedies, were specifically excluded.

Magnet samples were purchased from two specified domestic commercial producers. We also made some measurements on samples from a few other domestic and foreign sources as time permitted. Magnets for this purpose were provided free by a number of cooperating companies. A complete outline of the planned investigation and the organization of the effort was given in the two preceeding semi-annual progress reports.[1,2] These reports should also be consulted for details concerning the instrumentation and measurement techniques employed and a discussion of special problems in the acquisition and interpretation of the data presented herein.

The present report deals with the following subjects:

(a) The long-term stability of the open-circuit remanence at room temperature.

Magnet samples selected to represent the entire range of intrinsic coercive forces initially measured were stored individually, one group in

1. H. F. Mildrum and K. J. Strnat, "Research to Investigate the Aging Characteristics of Samarium Cobalt Magnets", AFML-TR-73-46, March 1973.

2. H. F. Mildrum and K. J. Strnat, "Research to Investigate the Aging Characteristics of Samarium Cobalt Magnets", AFML-TR-73-249, October 1973.

air and one in vacuum, and their remanent flux measured at regular time intervals with a high-resolution digital fluxmeter. The samples were partly in the as-received state, partly pre-stabilized by heating for 1 hour to 150°, 200° or 250°C. These tests extended over 7000 to 8000 hours.

(b) The stability of open-circuit remanence during exposure to various elevated temperatures in air.

Magnet samples selected to represent the entire range of intrinsic coercive force were aged at the temperatures 150°, 200°, 250° and 300°C. At certain time intervals, the samples were cooled to room temperature, their open-circuit remanent induction flux measured, and then they were reheated to continue the test. Most samples were entered in the as-received condition, so that the initial irreversible loss could be evaluated, but some were pre-stabilized by heating to simulate the procedure used by most manufacturers before sale of the magnets It should be pointed out that we had specified that the samples should not be subjected to the pre-aging, or "thermal knockdown", treatment which is routinely applied by most producers with conditions depending upon the use of the magnets specified by the customer.

(c) The permanent part of the irreversible changes of magnetic properties during heat aging.

The elevated-temperature exposure tests were terminated after the remanence had dropped significantly below its prestabilized value. On these samples, demagnetization curves from the reduced-remanence state and complete hysteresis loops after remagnetization were measured. Such curves reveal changes in the intrinsic magnetic properties and allow a differentiation between the purely magnetic and the structurally-caused ("metallurgical") parts of the irreversible losses.

(d) The changes of the open-circuit remanent induction flux during repeated thermal cycling of magnets between room temperature and 300°C.

These experiments were performed on axially magnetized cylindrical samples of 1/4" diameter with dimensional ratios corresponding to an approximate open-circuit unit permeance of B/H = 1/8, 1/4, 1 and 2-1/2.

The results of these measurements permitted the computation of the so-called irreversible and reversible losses, and of reversible temperature coefficients.

(e) Compressive strength of the structural magnet body.

Magnet samples representing the entire range of intrinsic coercive force were subjected to compressive loading until evidence of structural failure was observed. These experiments were performed from room temperature to $250^{\circ}C$.

(f) Aging under compressive load.

Long term tests were also conducted on samples compressively stress-loaded with 10,000 psi and heated at temperatures between 150° to $250^{\circ}C$. The samples had been prescreened at $25^{\circ}C$ to withstand this static compressive load. Results are compared with those of elevated temperature air aging experiments done without mechanical loading.

(g) Surface material removal by surface grinding and electric spark discharge machining.

The effects of material removal by standard centerless grinding and electrical discharge machining on the magnetic properties and their stability were investigated. Large diameter samples (0.500 dia. B/H = 1, 1/8) were reduced in diameter to 0.250 inches (B/H = 2-1/2 and 1/4) by each method and at two different machining rates. This was done with samples from both suppliers. Additional machining by surface grinding was performed on Raytheon samples (B/H = 1) to reduce them to B/H = 1/4 since no such thin samples had been supplied initially.

Samples thus machined were subsequently thermally aged at temperatures of 150° to $250^{\circ}C$ for comparison with samples aged from the "as-received" condition.

(h) Effects of alternating magnetic field effects on the stability of the open-circuit remanent flux operating point.

Several identical samples of each manufacturer were placed in the closed circuit of an iron-yoke electromagnet and subjected to a cyclic field varying by $\pm$ 0.86 H_d symmetrically about H_d. At certain time intervals the cyclic fields were reduced to zero, and the open-circuit remanent flux measured.

(i) Correlation between manufacturing conditions, magnetic properties and aging behavior.

We requested from the magnet manufacturers a detailed description of the general manufacturing processes used and of the specific conditions which applied to the various lots of samples we tested. Considerable detail was indeed supplied and much applicable general information can also be found in the reports written by the Raytheon and General Electric Companies while their manufacturing processes were being developed under US Air Force contracts.[3,4] This processing information was summarized in a previous progress report.[2]

General comments are made where possible, relating magnet processing variations to specific aging results.

3. M. G. Benz, et al., "Manufacturing Methods and Technology for Processing Cobalt-Samarium Magnets", AFML-TR-71-142, July 1971.

4. D. Das, et al., "Manufacturing Methods for Samarium Cobalt Magnets", AFML-TR-71-151, August 1971.

SECTION II

SAMARIUM COBALT MAGNET EVALUATIONS

1. EVALUATION OF THE LONG-TERM STABILITY AT ROOM TEMPERATURE

To determine the "natural" aging characteristics at room temperature of $SmCo_5$ magnets, samples from each producer were selected such that the extreme values of ${}_MH_c$ and in some cases also of H_k, observed within each lot were reasonably well represented. This also provides a reasonably large spread of values. In each case a hysteresis loop was measured first and the test samples were then recharged in a 38.8 kOe static field prior to the initiation of the aging test.

Most of these tests were terminated at the end of the contract period. The final results of these evaluations are presented in Tables 1 and 2. In Table 1, the open-circuit remanent induction (OCRI) values measured on samples having a permeance $B/H \approx 1/8$, 1/4 and 1 which were stored in air at room temperature under the normal atmospheric conditions of our laboratory are shown as a function of time. ("Natural air aging"). Generally speaking, the OCRI loss after about 7000 hours is in the range of 0.20 - 0.98% for both sample lots ($B/H \approx 1$). It appears that the die-pressed magnets show consistently greater losses than the isostatically pressed ones. They also had lower H_k, i.e., less "square" demagnetization curves than the isopressed magnets. It seems that H_k is the one magnetic property that can be related most directly to the causes of aging. In the present set of data, there is no obvious correlation of the losses with the magnitude of ${}_MH_c$. The range from best to worst-case observed aging behavior is shown in Figure 1.

Table 1 also contains the natural air aging results for shorter length samples having a $B/H \approx 1/4$ ($L/D = 0.10$) and $B/H \approx 1/8$ ($L/D = 0.044$). It can be seen that even during the short duration (3000 hours) of the tests on these thinner samples, the same or higher losses are incurred as in twice the time for the thicker samples. Thus there is a significant increase in the rate of flux loss with decreasing thickness. Since these thinner samples have increasingly higher surface area-to-volume ratio, this finding suggests that oxidation involving the diffusion of oxygen from the outer surface plays a significant role in the natural aging.

Another set of 4 samples with very similar initial properties ($B/H \approx 1$ group) underwent room-temperature aging in a vacuum of about 1×10^{-4} Torr from which they were periodically briefly removed for measurement purposes. The results of this experiment are shown in Table 2. The level of losses in this experiment was very similar to that of comparable samples kept in air. However, the level of losses incurred by one of the air aged die-pressed samples increased significantly after ~3500 hours while the other was comparable to its vacuum-aged counterpart. It must be concluded that the periodically interrupted vacuum storage had little protective value at room temperature. See Figure 1, worst case "as-received".

To determine if room-temperature aging effects can be minimized by a thermal stabilization treatment, several samples were given a specific time-temperature "knockdown" prior to initiating the room-temperature storage test. The first group of samples so tested were selected from the first and second lots purchased from each producer and represented ${}_{M}H_{c}$ values near the average and upper extreme observed in each lot. A thermal knockdown exposure of 250°C for 1 hour was first performed. After cooling to room temperature, OCRI measurements were conducted on a routine schedule. The data for these samples (Table 3, lines 1-4) indicates for the most part an increased stability against natural air aging in comparison with magnet samples of similar properties listed in Table 1. Perhaps significantly, the first-lot samples from each manufacture (A-13, B-11) show little or no additional aging beyond the "knockdown even after ~8000 hours, whereas second-lot samples (A-33, B-40) suffered an additional loss of ~0.2% at the end of ~7000 hours.

A few samples having average magnetic properties were subsequently selected from the second lot of each manufacture. (A-35 through B-42).

TABLE 1

NATURAL AGING IN AIR AT ROOM TEMPERATURE

D = 0.25 in. L = 0.1 in (L/D = 0.4, B/H ≈ 1)

Sample Number	Total Elapsed Time (Hours)	Total Loss (%)	Initial Properties $_MH_c$ (kOe)	Initial Properties H_k (kOe)	Lot Number
A-11	8000	0.35*	20.3	8.20	A-1
A-47	7000	0.20	44.3	9.91	A-2
B-18	8000	0.98	36.8	5.30	B-1
B-37	7000	0.51	33.2	6.63	B-2
D = 0.25 in, L = 0.025 (L/D = 0.1, B/H = 1/4)					
A-200	3000	0.37	25.4	9.0	A-2
A-207	"	0.18	25.0	7.2	A-2
A-214	"	0.19	34.8	5.4	A-2
B-204	"	0.80	25.4	2.8	B-2
B-210	"	0.78	23.7	2.9	B-2
B-211	"	0.66	26.5	3.0	B-2
D = 0.25 in, L = 0.011 (L/D = 0.044, B/H = 1/8**)					
A-400	3000	0.52	31.9	3.6	A-2
A-401	"	0.64	28.5	3.5	A-2
A-402	"	0.53	25.0	3.4	A-2

*Sample chipped after 3200 hours, subsequent OCRI data corrected for difference in value before and after material loss

** B-samples of B/H = 1/8 not available. See text Section IV.

TABLE 2

AGING IN VACUUM AT ROOM TEMPERATURE

D = 0.25 in, L = 0.1 in (L/D = 0.4 , B/H ≈ 1)

Sample Number	Total Elapsed Time (Hours)	Total Loss (%)	Initial Properties $_MH_c$ (kOe)	Initial Properties H_k (kOe)	Lot Number
A-20	8000	0.25	20.2	8.95	A-1
A-46	7000	0.25	44.8	8.12	A-2
B-17	8000	0.53	36.4	4.97	B-1
B-35	7000	0.52	34.5	8.09	B-2

TABLE 3

NATURAL AIR AGING AFTER A STABILIZATION HEAT TREATMENT OF ONE HOUR AT VARIOUS TEMPERATURES *

("THERMAL KNOCKDOWN")

Sample Number	Treatment Temperature (°C)	Initial Loss (%)	Total Elapsed Time-Hours	Additional Loss (%)	Initial Properties $_MH_c$ (kOe)	Initial Properties H_k (kOe)	Lot Number
A-13	250	16.51	7885	0.06	18.1	8.84	A-1
A-33	250	4.60	6989	0.21	46.2	9.44	A-2
B-11	250	5.33	7885	0.00	35.7	4.39	B-1
B-40	250	7.80	6989	0.20	33.1	6.35	B-2
A-35	150	2.25	4276	0.05	43.0	7.79	A-2
B-41	150	4.40	4276	0.12	36.6	6.21	B-2
A-45	200	3.11	4276	0.05	41.9	8.01	A-2
B-42	200	4.54	4276	0.12	36.3	7.84	B-2

* $B/H \approx 1$, Sample Diameter 1/4"

TABLE 4

COMPARISON OF NATURAL AIR AGING OF MAGNETS FROM THE AS-RECEIVED CONDITION AND FROM DIFFERENT THERMALLY PRESTABILIZED STATES

Prestab. Treat.	Initial Loss* (%)	Additional Loss** (%)	Evaluation Time (Hours)
none	none	0.18 - 0.4	4200
(1 hr, 150°)	2.2 - 4.4	0.05 - 0.12	4200
(1 hr, 200°)	3.1 - 4.5	0.05 - 0.12	4200
(1 hr, 250°)	4.6 - 7.8	0.00 - 0.21	4200

* In the case of stabilized samples the initial loss represents that incurred during the prestabilization heat treatment.

** Loss during subsequent aging; % of reduced, stabilized value.

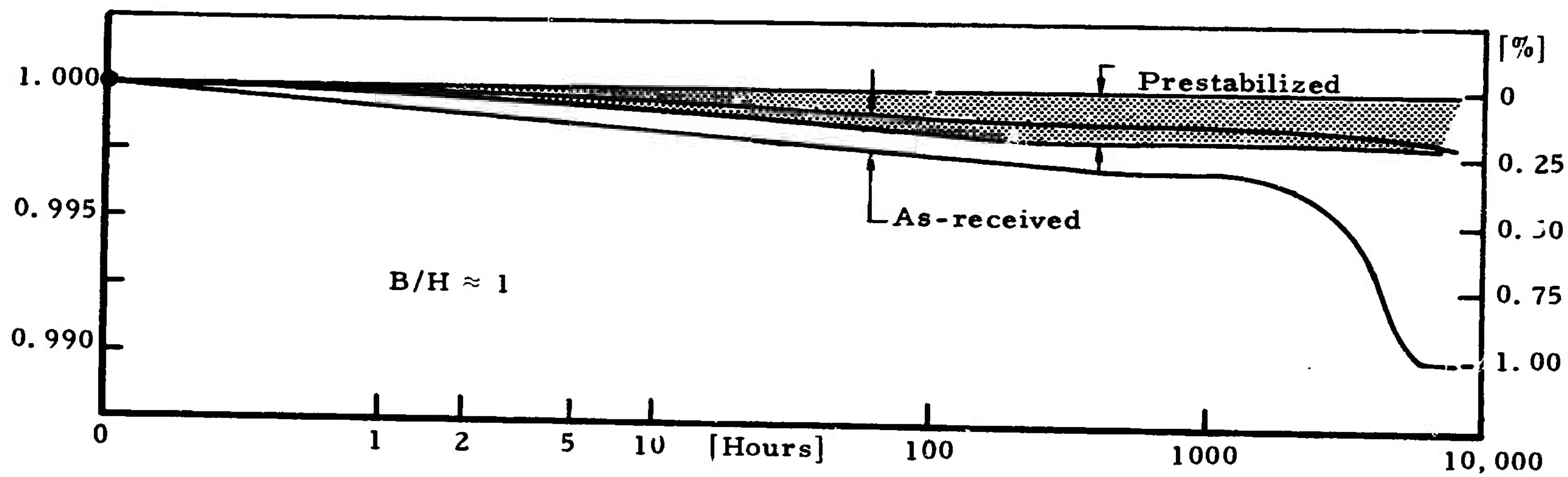

Figure 1. Natural Air Aging at Room Temperature. Comparison of Best and Worst Case Normalized Curves. For Sampes "As-Received" and Prestabilized. B/H ≈ 1.

They were similarly prestabilized, but at the lower temperatures of 150° and 200°C. After approximately 4000 hours evaluation time a comparison indicates that even a lower thermal knockdown treatment level improves the natural aging stability. The isopressed samples fared generally better after this lower-temperature stabilization while the diepressed sample, B-11, stood out as highly stable after the 250° knockdown.

It must also be pointed out that this knockdown treatment understandably is not without its undesirable consequences. As noted in Table 3, the level of treatment incurs a moderate to substantial initial loss of open-circuit remanent induction ranging from 2.2 - 16.5 percent.

A more direct comparison of the merits of thermal prestabilization of these magnets for ambient applications is shown in Table 4. Here we present data on sample magnets from the second lots only, of both manufacturers (no distinction made), for an identical aging interval of 4200 hours for all samples.

2. LONG-TERM AGING TESTS IN AIR AT ELEVATED TEMPERATURES

The basic objective of these tests was to evaluate $SmCo_5$ magnets for applications in devices in which they are used to produce a static magnetic field, operate on a loadline of B/H = 1 or less, and where they are exposed to elevated temperatures in normal atmosphere for prolonged periods of time under operational, manufacturing or storage conditions. The test temperatures were selected primarily with a view toward microwave tubes where Sm-Co magnets have found their first extensive application. Traveling wave tubes are now built to operate up to about 150°C, advanced designs to perhaps 200°C, with single magnets operating in hot spots that may go even higher. Tube builders would like to design even hotter tubes, and we were informed that 300°C was a reasonable upper limit.

Samples for these tests were selected from the lots of both producers as previously described.[1,2] The sample shape selected for the initial test series was 1/4" diameter and 1/10" thickness, corresponding to B/H ≈ 1. It was decided to begin the exposure tests at the upper temperature bound of the range of interest, 300°C, because one would expect the fastest aging at this highest temperature. The entire range of phenomena should thus be seen in the shortest possible time. We wanted to nearly finish the "aging runs" on a few initial samples before engaging in a massive routine-testing program. This procedure allowed a critical evaluation and improvement of the experimental methods chosen and a more prudent selection of the time intervals for data-taking which would considerably reduce the effort involved in conducting subsequent tests.

Presentation of the data in this chapter is such that only a tabular numerical summary of the main results of the tests at each temperature is given. Some selected normalized curves, which allow one to see general trends, also appear in the text. Detailed graphical presentations of the aging behavior of many samples for exposure times up to 4200 hours were printed in the second report.

In this report we have redefined the initial loss as the loss incurred during the first 15 minutes of aging at given exposure conditions. Additional losses incurred are also referred to this time reference. Normalization for those curve sheets comparing the range from best to worst cases for a given evaluation are also made with the same reference, the flux value after 15-minute exposure, except where specifically noted.

For the preliminary data presentation in the preceding report, a reference point of 2 hours had been chosen on the assumption that it marked the (poorly defined) end of an initial period of rapid flux reduction at 300°C. However, for other temperatures of exposure, this period becomes increasingly longer as the temperature is chosen lower. Samples tested at 150°C without a prior "thermal knockdown" appear to be still in the period of protracted initial, irreversible loss even after several thousand hours of exposure. The 2-hour reference was thus meaningful only for the 300° tests, but not at any lower temperature.

We therefore abandoned any attempt to define the onset of the plateau of temporary stability in a general way and decided to use the first point of measurement after exposure began - usually 15 minutes - as the one to which all flux values should be referenced for the purpose of making generalized comparisons of aging curves.

a. Air Aging Tests at 300°C.

The results of these are shown in Table 5. The upper half of the table shows the changes of the open-circuit remanent induction (OCRI) of magnets which were recharged in the hysteresigraph yoke with a 38.8 kOe d. c. field after plotting of the initial hysteresis loop. The lower half of the table refers to samples recharged in a 75 kOe field of a superconducting magnet at the Air Force Materials Laboratory. For each sample, selected initial magnetic properties ($_MH_c$, H_k) are listed to permit correlations with the aging behavior.

The "initial loss" is the reduction from the initial room-temperature value of the OCRI during the first 15 minutes of accumulated exposure time. For most magnets a noticeable additional reduction was seen before something of a plateau was reached. In the second column of the table, this initial loss is expressed in percent of the starting value (before heating) of OCRI. The "additional loss" listed in the third column is referred to the prestabilized OCRI value after 15 minutes exposure. The same is true for the 1%, 3% and 5% loss specified at the head of the table, columns 5, 6 and 7 respectively. For each of the % loss figures, the corresponding total elapsed exposure time is given, which can be considered a kind of "lifetime" of the magnet provided the user can tolerate the specified loss of useful flux.

As the 300°C evaluation tests progressed, and time and oven space became available, a single sample (D-4) furnished by the Electron Energy Corporation was also included. Samples furnished by this producer (our letter code D) were diepressed. They had the standard dimensions. At our request, these samples were also furnished in an unstabilized condition. Open-circuit remanence of this sample was measured in the same manner and time pattern as for the other producers' samples.

In the first several tests, the exposure times were extended far beyond the point which would normally be of practical interest, namely until the additional loss reached values between 10 and 20%. (See Figure 2) This was done to see if there might be a restabilization at a secondary, lower level where the OCRI is still high enough to be useful. Such a restabilization did not occur for the GE and Raytheon test magnets. For these samples, at 300°C in air, a 1% loss is incurred in the first 5-25 hours of exposure, 3% loss was observed after 80-225 hours, and 5% after 200-750 hours. Then the rate of the drop becomes progressively greater and the decay toward the end of the extended tests must be considered catastrophic. Some indication of a correlation between the magnitude of ${}_{M}H_{c}$ and the loss rate can be seen. It appears that magnets with higher ${}_{M}H_{c}$ values have a somewhat longer "lifetime" at this temperature than those with lower initial coercive force. This seems to be equally true for magnets from each of the two producers.

However, the single magnet from the Electron Energy Corporation exhibited aging characteristics markedly different from those observed on samples from our two primary sources of test magnets. As shown in Table 5, this particular magnet has a substantial lower ${}_{M}H_{c}$ value and a higher H_k value - hence a greater "squareness" of the demagnetization curve. Its initial loss of 4.7% is on the low end of the scale, and although the additional loss to 1% is ~36 hours, the total loss for ~3500 hours is only 2.73% - very much less for any other sample tested. We have taken the liberty of estimating a 3% loss at approximately 10,000 hours based on projecting the slope of the curve between 200-3500 hours. Unlike the other magnets tested at 300°C, this sample reached a second plateau after several 100 hours accumulated aging time and no really catastrophic decay had yet set in at the end of the test, after ~3500 hours.

In the lower portion of Table 5, we show results for those magnets which were recharged in a superconducting solenoid field of 75 kOe. Surprisingly, these were observed to have an even shorter life span than samples charged at our standard value of 38.8 kOe. As one would expect,

TABLE 5

ACCELERATED AGING IN AIR AT 300°C, B/H ≈ 1

Sample Number	Initial Loss* (%)	Total Additional Loss: Loss** (%)	Total Additional Loss: Time (Hours)	Time to Loss of: 1% (Hours)	Time to Loss of: 3% (Hours)	Time to Loss of: 5% (Hours)	Initial Properties: $_MH_c$ (kOe)	Initial Properties: H_k (kOe)	Lot Number
A-25	5.6	10.4	3615	24	225	750	48.9	8.48	A-2
A-31	6.2	15.7	1616	20	110	275	32.5	9.03	A-2
B-27	10.7	19.4	1616	5	80	200	34.1	5.52	B-2
B-31	8.9	12.2	3615	22	168	400	39.2	6.43	B-2
D-4	4.6	2.73	3473	36	~10,000***	---	23.7	12.8	---
AGED AFTER CHARGING TO 75 kOe									
A-32	3.8	10.3	2255	15	75	250	47.7	7.81	A-2
A-40	8.4	33.6	1253	15	70	130	37.1	6.63	A-2
B-29	8.5	13.0	2255	25	150	325	38.8	6.05	B-2
B-43	10.8	11.6	2255	10	75	200	29.6	5.96	B-2

Note: All magnets had the dimensions D = 0.25", L = 0.1".

* Drop during first 15 minutes, expressed in % of initial value

** Drop from the value at 15 minutes, expressed in % of this value

*** Estimated

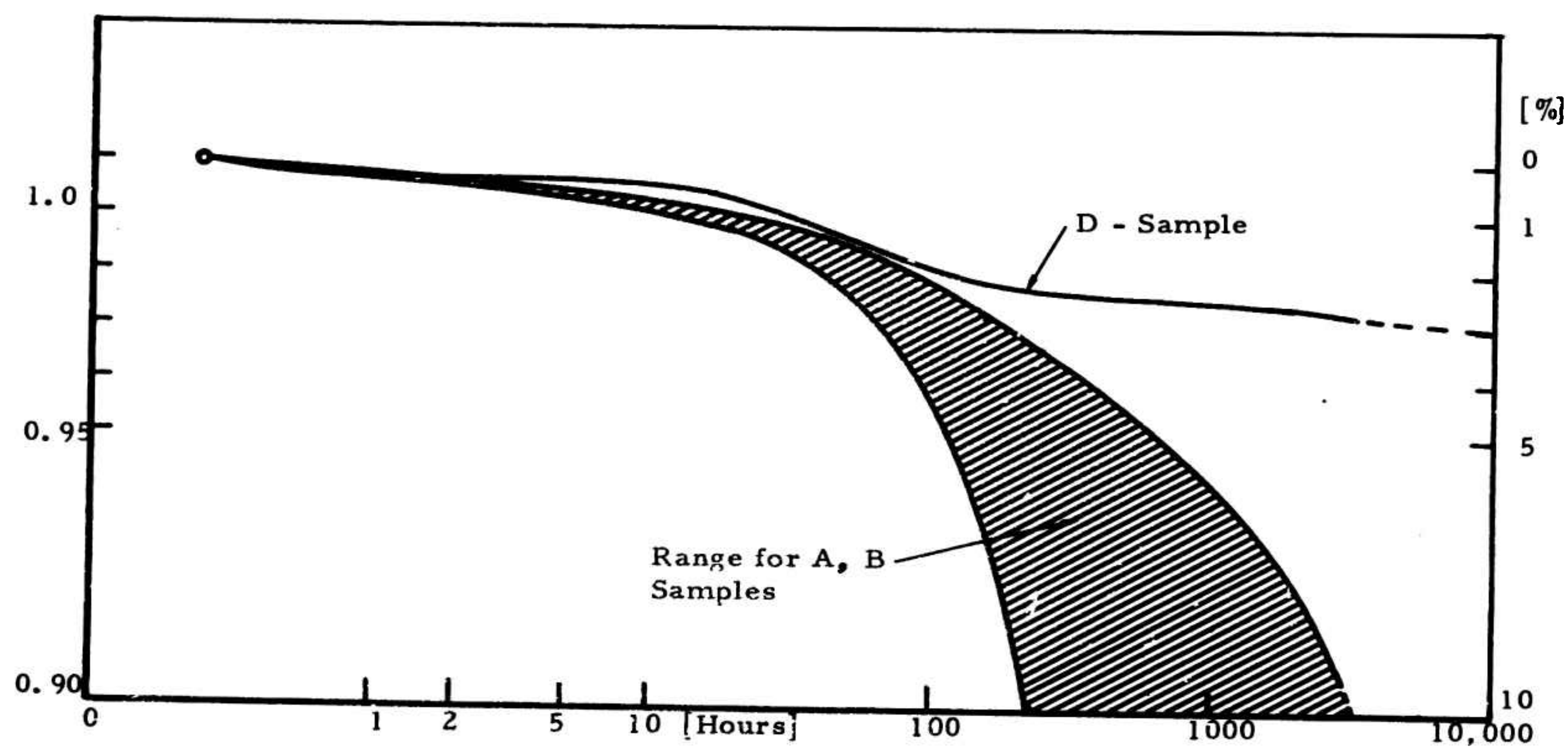

Figure 2. Air Aging at 300°C. Comparison of Best and Worst Case Normalized Curves. B/H ≈ 1.

the initial OCRI values were slightly higher indicating a better approach to saturation, and in most cases the OCRI at the 15-minute reference point were higher, too. But aging times to a loss of 1, 3 or 5% were shortened substantially so for the isopressed magnets (code A). The correlation with high and low $_MH_c$ values still exists. Coercivity and H_k were not significantly increased by the higher peak magnetizing field. These results appear to indicate that very-high-field charging of a magnet does nothing to enhance its aging characteristic. It must be recognized, however, that we could not use sufficiently large numbers of samples in these tests to make the results statistically meaningful.

Generally speaking, the stability at 300°C was disappointingly poor, based on our experimental results for magnets produced by GE and Raytheon in mid-1972. At this point in time, the use of $SmCo_5$ magnets at this operating temperature can therefore not be generally recommended. However, for applications where the thermal exposure is of very short duration, or for some one-time applications in which reasonable losses can be tolerated, these magnets would suffice. We have since been assured by personnel of both producers (Hitachi Magnetics as the successor of GE, and Raytheon) that certain specific production changes have recently been made which improved the elevated-temperature stability of their products. Additional aging studied on more recent production samples must be made to prove claims of improved performance. If the fairly stable behavior observed on sample D-4 is taken as an indication of the potential stability attainable for $SmCo_5$ magnets, then there is obviously room for substantial improvement of diepressed magnets in this respect. It must be concluded that magents that will perform satisfactorily for a long time at 300°C are a definite practical possibility.

b. Air Aging Tests at 250°C

A somewhat larger group of samples was chosen for aging tests at 250°C: from the two General Electric lots, including one (A-39) which had a very uncharacteristically severe step in the demagnetization curve near field zero, and from the two Raytheon lots. Samples of $B/H \approx 1$, 1/4, 1/8, and 2-1/2 were eventually exposed to the 250°C for evaluation. In this section, only samples aged from the "as-received" state will be discussed. A comparison will later be made in the section dealing with the aging performance of magnets machined to proper length-to-diameter ratios.

In this section we have again included the results on a single sample, (D-3), obtained from the Electron Energy Corporation. Isopressed, liquid-phase-sintered magnet samples produced by Brown, Boveri & Cie were also evaluated (150 - 250°C) over 3000 hours. These results were not tabulated. The observed performance was quite similar to that of magnets of GE production at approximately the same time.

For the A and B sample groups, the samples were selected to represent the whole range of intrinsic coercive force values. As before, most of the magnets were charged in a 38.8 kOe steady field, but four additional ones were magnetized with 75 kOe.

The results for 1/4" diameter samples of $B/H \approx 1$ unit permeance are presented in Table 6, and the best-worst cases illustrated in Figure 3. The following general conclusions may be drawn from the data: The stability at 250°C is significantly greater than at 300°C; for the best samples of either manufacture the increase in lifetime is 10 to 30 fold, but for the worst cases the improvement factor is only 2 to 3. Extremely large variations in "quality" were observed within each of the two major groups of samples; there is no indication that either the isostatic pressing or the diepressing method produces inherently more stable samples. Within each group, the samples of the initial lots (A-1 and B-1) were more stable than those of lots A-2 and B-2 received later. One is tempted to conclude that, either, the changes in the production process during the early "tooling-up" stages reduced the stability of the magnets, or that the magnets of the small initial lots were pre-screened by the manufacturers even though it had been agreed that this should not be done. (Although it must again be emphasized that our conclusions are drawn from sample quantities too small to be

TABLE 6

ACCELERATED AGING IN AIR AT 250°C, B/H ≈ 1

Sample Number	Initial Loss* (%)	Total Additional Loss: Loss** (%)	Total Additional Loss: Time (Hours)	Time to Loss of 1% (Hours)	Time to Loss of 3% (Hours)	Time to Loss of 5% (Hours)	Initial Properties $_M H_c$ (kOe)	Initial Properties H_k (kOe)	Lot Number
A-16	11.5	5.8	5023	8	1300	3650	22.9	12.23	A-1
A-18	8.24	5.1	5023	175	2975	4680	18.5	6.82	A-1
A-21	4.2	5.0	4158	75	1570	4158	33.0	11.32	A-2
A-39***	6.9	18.7	2159	22	200	270	37.5	1.85	A-2
A-44	5.2	10.53	2159	16	220	525	49.6	5.47	A-2
B-12	4.8	3.0	7672	200	7672	----	36.0	4.83	B-1
B-20	5.0	2.6	7672	900	----	----	34.2	3.95	B-1
B-32	12.1	7.2	4158	1	350	1550	22.4	4.53	B-2
B-45	6.1	4.7	4158	4	1700	----	42.3	8.09	B-2
D-3	3.8	1.3	3473	1100	----	----	25.5	9.70	---
AGED AFTER CHARGING TO 75 kOe									
A-22	4.1	3.9	4390	54	1850	----	48.8	7.50	A-2
A-29	3.8	5.6	2413	4	554	2080	40.6	7.01	A-2
B-25	5.3	4.9	4390	25	1900	----	39.8	6.60	B-2
B-47	7.7	6.1	4390	10	1500	3750	31.6	5.63	B-2

* Drop in first 15 minutes, expressed in % of initial value

** Drop from the value at 15 minutes, expressed in % of this value

*** Not a representative sample

NOTE: Sample dimensions are D = 0.25", L = 0.1" (L/D - 0.4, B/H ≈ 1)

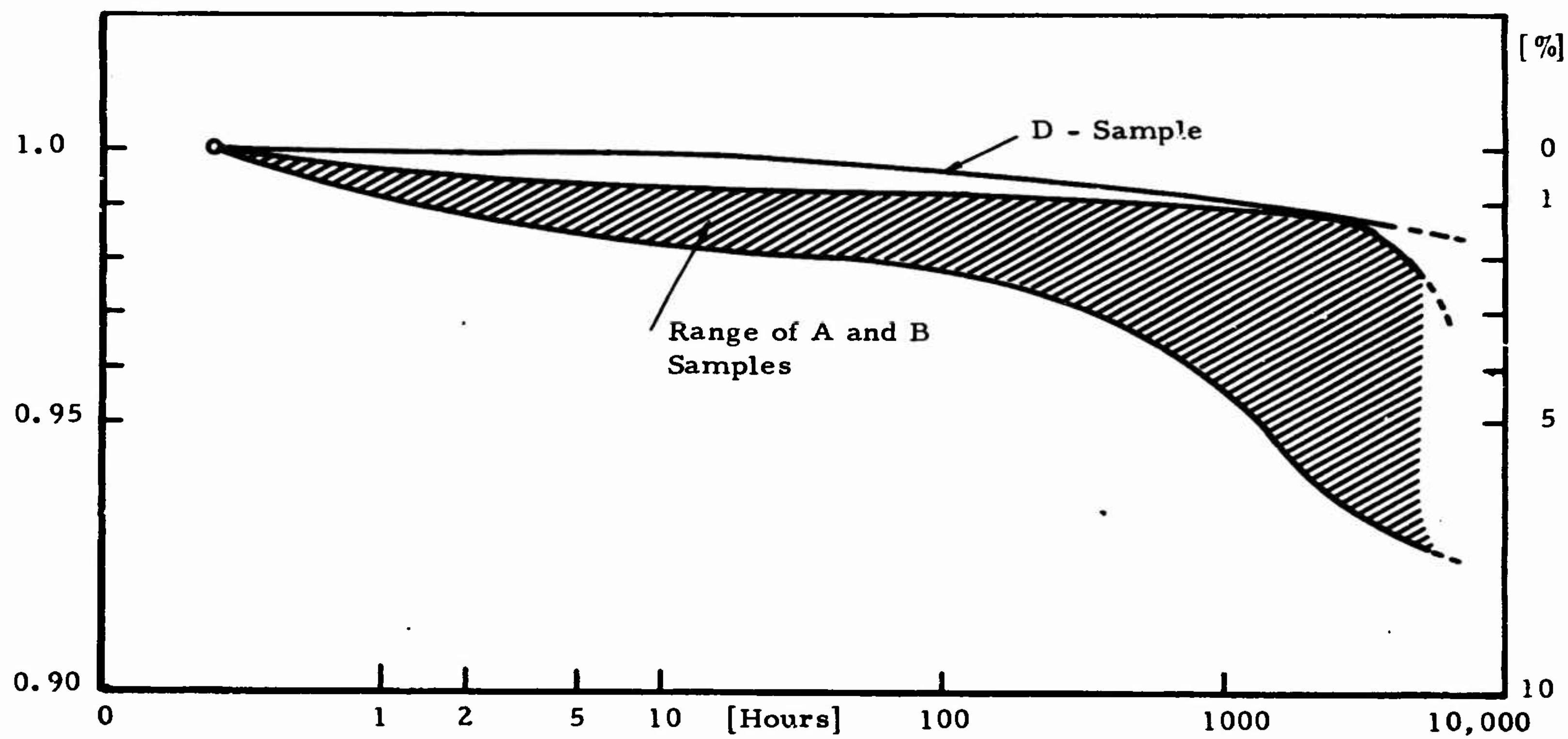

Figure 3. Air Aging at 250°C. Comparison of Best and Worst Case Normalized Curves. B/H ≈ 1.

representative of a given product in a statistical sense.)

No clear correlations can be seen between the aging stability, as measured by the "time to loss of 1%, 3%, 5%," and either the initial values of $_MH_c$ and H_k, or the "initial loss" during the first 15 minutes of heating. No exceptional behavior could be observed for the magnets that had been pulse charged at 75 kOe.

In Table 7 we present 250° aging results on samples received from GE which had $B/H \approx 1/4$ and 1/8 unit permeance. Raytheon declined to furnish the samples needed - - 0.250" diameter, 0.025" and 0.012" thick, respectively - - and therefore representative B samples do not appear in this table. The samples selected for this evaluation were matched in pairs having almost identical B_r, $_MH_c$, and H_k properties representative of the range of samples with this geometry.

The data presented in the table and illustrated in Figure 4 a and b show initial losses of 11-18% for samples having $B/H \approx 1/4$, and 11-16% loss for $B/H \approx 1/8$ samples. On the average, these initial loss values are considerably higher than those incurred by A-group samples of $B/H \approx 1$ under the same exposure. This can be qualitatively understood, as was pointed out by K. Bachmann[5], when one considers the difference between the demagnetizing field and the intrinsic coercive force, $_MH_c$ (or perhaps better, the "knee field", H_k) at the exposure temperature as the principal factor controlling the short-term, "irreversible" losses. These thin samples have considerably higher self-demagnetizing fields than the $B/H \approx 1$ samples, and thermally activated domain-wall motion which reduces the remanence is thus much more likely to occur at 250°. It should also be noted that the H_k values for these samples of $B/H \approx 1/4$ and 1/8 are generally lower (3.3-4.5 kOe) than those of the type A reference samples of $B/H \approx 1$ (5.5-12.2 kOe, with one exception, H_k = 1.85 for A-39!), a fact which would favor the partial thermal self-demagnetization of the thin samples.

5. K. Bachmann, "Reversible and Irreversible Losses of Magnetization in $SmCo_5$ Magnets". Paper No. 6E-7, 19th Conf. on Magnetism and Magnetic Materials, Boston, Mass. Nov. 1973.

TABLE 7

ACCELERATED AGING IN AIR AT 250°C, B/H = 1/8, 1/4
(As Received Lot A-2)

Sample Number	Initial Loss* (%)	Total Additional Loss Loss** (%)	Total Additional Loss Time (Hours)	Time to Loss of 1% (Hours)	Time to Loss of 3% (Hours)	Initial Properties $_M H_c$ (kOe)	Initial Properties H_k (kOe)	B/H
A-218	12.5	3.0	1879	3	653	24.33	4.53	1/4
A-221	17.9	3.3	1879	1	620	24.48	4.20	1/4
A-222	12.6	1.3	1879	320	---	28.95	4.20	1/4
A-253	11.0	1.5	1879	150	---	27.42	4.65	1/4
A-422	14.6	5.0	1879	0.5	190	24.30	3.60	1/8
A-420	16.0	5.5	1879	0.5	190	24.36	3.30	1/8
A-414	14.0	3.9	1879	0.5	850	26.16	3.60	1/8
A-405	11.2	3.3	1879	1.5	1000	26.37	4.14	1/8

* Drop during first 15 minutes, expressed in % of initial value.
** Drop from the value at 15 minutes, expressed in % of this value.
NOTE: All samples had dimensions D = 0.25"

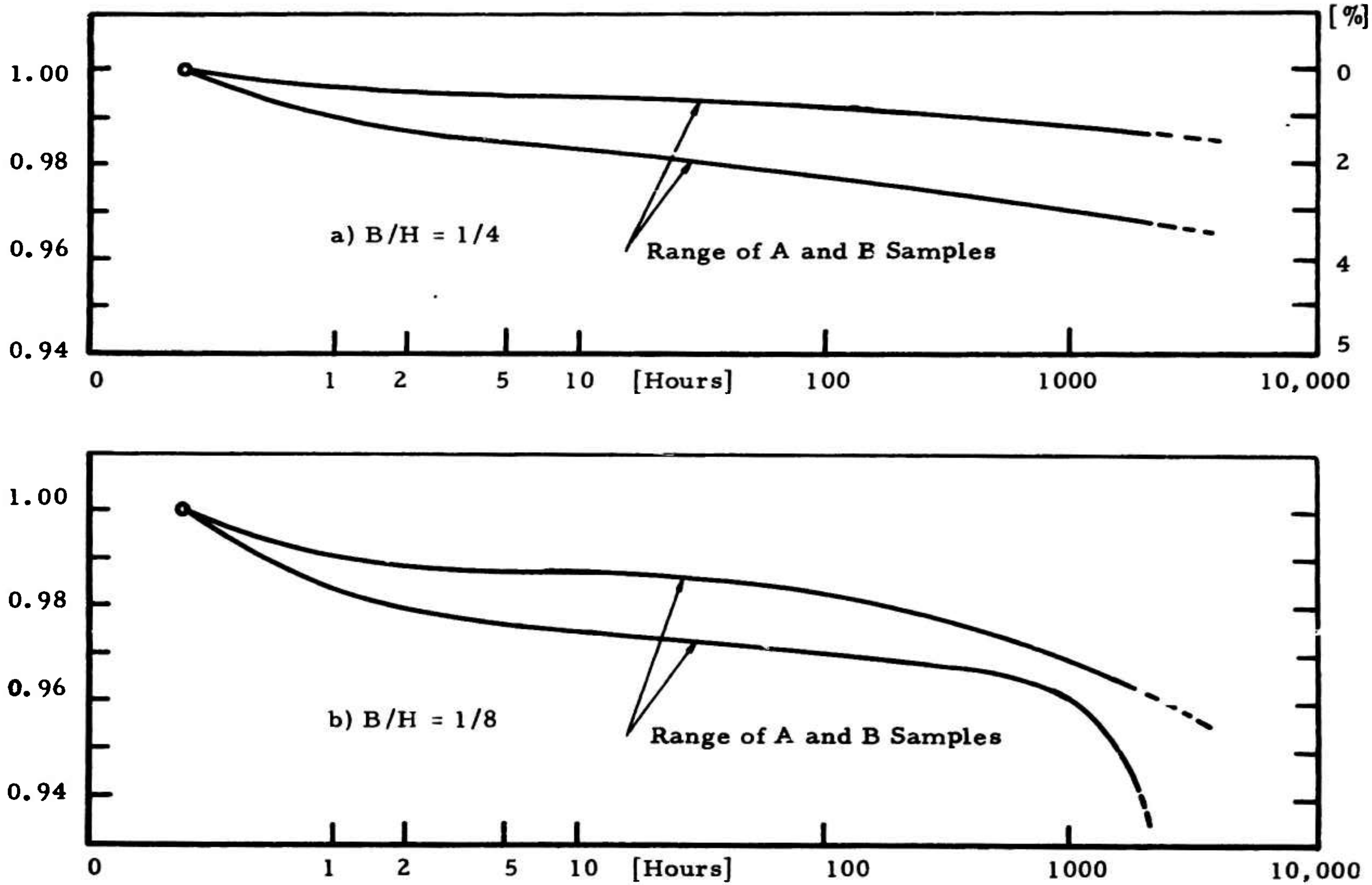

Figure 4. Air Aging at 250°C. Comparison of Best and Worst Case Normalized Curves.
a) B/H = 1/4 b) B/H = 1/8

The values of the elapsed aging time to an additional loss of 1 and 3% for these samples are informative due to the similarity in the matched pairs with low and high ${}_{M}H_{c}$ for both values of unit permeance. Here we see evidence that a 1% additional loss occurs much sooner for the samples of smaller thickness (B/H ≈ 1/8) than for the thicker ones of B/H ≈ 1/4. Also the time for a loss of 3% appears to show a reasonably clear correlation to the magnitude of ${}_{M}H_{c}$. That is, the higher ${}_{M}H_{c}$ the longer the period for the additional loss to reach the 3% level.

While the initial and short-term aging losses are likely to be due to purely magnetic causes (thermally activated wall motion and/or domain nucleation), the long-term aging loss could well be attributable to a considerable extent to structural changes of a metallurgical nature or to oxidation proceeding from the sample surface. If such oxidation from the surrounding atmosphere were an important factor, thin samples with a larger surface-to-volume ratio should show larger long-term losses. Such a trend seems indeed to be indicated by the figures in the "total additional loss" column of Table 7, with the loss ranging from 1.3 to 3.3 for B/H ≈ 1/4, and from 3.3 to 5.5 for B/H = 1/8. The figures for B/H = 1 samples in Table 6 are not consistent with this, but one must consider that the samples used there and their aging conditions were not matched to those dealt with in Table 7. Both, initial magnetic properties and total aging times for the B/H ≈ 1 magnets, covered a very wide range there, while properties were similar and aging times the same for all samples of Table 7.

c) Air Aging Tests at 200°C

At the culmination of about 4000 hours of air aging samples from the first and second lots of both producers at 200°C, it becomes apparent that exposure at this temperature level is generally not nearly as severe as that at 250° to 300° with regard to the magnetic stability of these magnets. This seems true regardless of manufacturing process and sample origin. Results are shown in Table 8. The initial-loss figures, ranging from 2.4 to 5.4%

are substantially lower than at 250°, as one would expect. But with regard to long-term aging, different individual samples showed quite different general behavior patterns. The two isopressed samples of the second GE lot, A-62 and A-70, aged at first at a slower rate than the other four A and B samples, but after a few 100 hours they entered the catastrophic decay phase generally characteristic of higher aging temperatures. Sample A-19 (first GE lot) and all B samples (Raytheon) aged at a higher initial rate; the remanent flux never truly stabilized during the test period, but there was also no indication of a catastrophic deterioration of the magnetic properties. Figure 5 shows aging curves characteristic for these two behavior types. Because of the crossing over of the lines, it was not possible to define a band within which all measured curves fall - as was done for other aging temperatures in any meaningful way.

An additional sample (D-2) of Electron Energy Corporation manufacture was included in this evaluation. It showed again superior stability. The initial, irreversible loss was the lowest at 2.4%, and the additional loss after approximately 3500 hours was only 0.5%. Again the squareness of the demagnetization loop as indicated by a high field value of H_k may be significant, although there were two other samples (A-19 and B-48) with even higher H_k which aged more severely.

In addition to evaluating these samples with a geometry giving $B/H \approx 1$ unit permeance, we also aged other samples from the "as-received" condition which had $B/H = 1/4$ and $1/8$. (Available only from G.E.). The results are shown in Table 9. As before, these samples were matched in pairs having magnetic properties as close as possible prior to the evaluation. They can be considered representative of the range of samples of that kind received by us.

The data for this evaluation is illustrated in Figure 6a, b. The initial loss now shows a clear increase with increasing demagnetizing field: 2.7 - 3.2% at $B/H \approx 1$, 8 - 9.5% at $B/H = 1/4$, and 10.5 - 16.8% at $B/H = 1/8$.

When one tries to compare numerically the aging losses at different

TABLE 8

ACCELERATED AGING IN AIR AT 200°C

$B/H \approx 1$

Sample Number	Initial Loss* (%)	Total Additional Loss: Loss** (%)	Total Additional Loss: Time (Hours)	Time to Loss of: 1% (Hours)	Time to Loss of: 3% (Hours)	Time to Loss of: 5% (Hours)	Initial Properties: $_MH_c$ (kOe)	Initial Properties: H_k (kOe)	Lot Number
A-19	2.7	3.0	3989	15	--	--	19.1	9.06	A-1
A-70	3.2	5.3	3989	58	1725	3150	37.0	3.04	A-2
A-62	2.9	6.2	3989	110	1080	2300	48.8	4.67	A-2
B-16	3.0	3.2	3989	5	1450	--	35.5	4.67	B-1
B-54	5.4	2.6	3989	6	--	--	29.4	6.07	B-2
B-48	3.3	2.4	3989	16	--	--	39.4	9.11	B-2
D-2	2.4	0.5	3473	--	--	--	29.4	8.6	--

* Drop during first 15 minutes, expressed in % of initial value

** Drop from the value at 15 minutes, expressed in % of this value

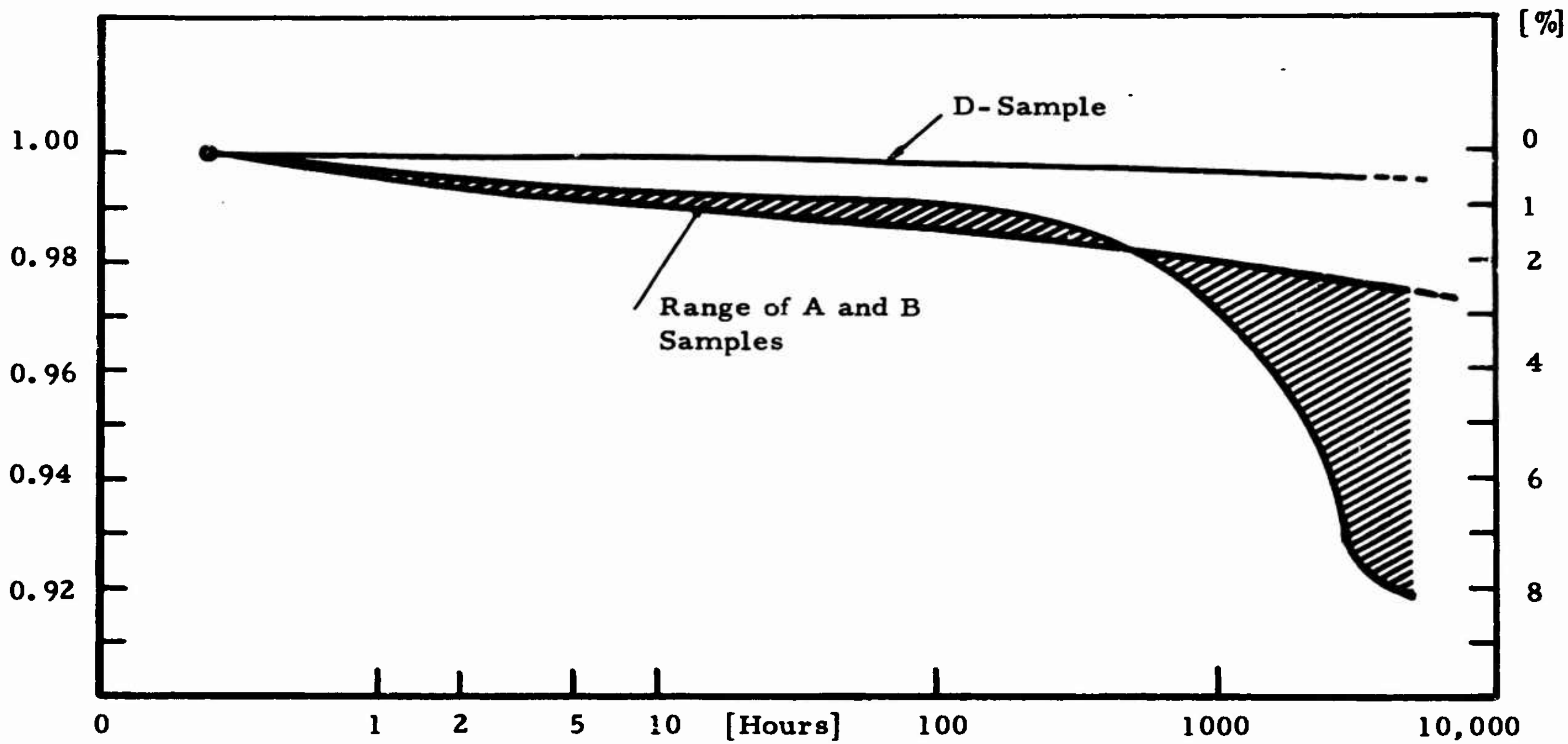

Figure 5. Air Aging at 200°C. Comparison of Best and Worst Case Normalized Curves. B/H ≈ 1.

TABLE 9

ACCELERATED AGING IN AIR AT 200°C, B/H = 1/8, 1/4
(As Received Lot A-2)

Sample Number	Initial Loss* (%)	Total Additional Loss Loss** (%)	Total Additional Loss Time (Hours)	Time to Loss of 1% (Hours)	Time to Loss of 3% (Hours)	Initial Properties $_M H_c$ (kOe)	Initial Properties H_k (kOe)	B/H
A-215	7.9	2.6	1842	5.0	----	24.09	4.95	1/4
A-233	7.9	2.7	1842	8.0	----	24.27	6.03	1/4
A-239	9.4	4.9	1842	0.8	7.0	26.04	7.35	1/4
A-240	8.4	3.9	1842	16.0	80	25.50	6.36	1/4
A-431	16.8	2.7	1842	4.5	----	24.81	3.66	1/8
A-438	17.2	2.8	947	2.4	***	25.26	3.66	1/8
A-423	11.2	4.2	1842	3.0	150	25.80	2.82	1/8
A-409	10.5	3.9	1842	2.9	150	25.53	2.82	1/8

* Drop during first 15 minutes, expressed in % of initial value
** Drop from the value at 15 minutes, expressed in % of this value
*** Sample cracked after measurement

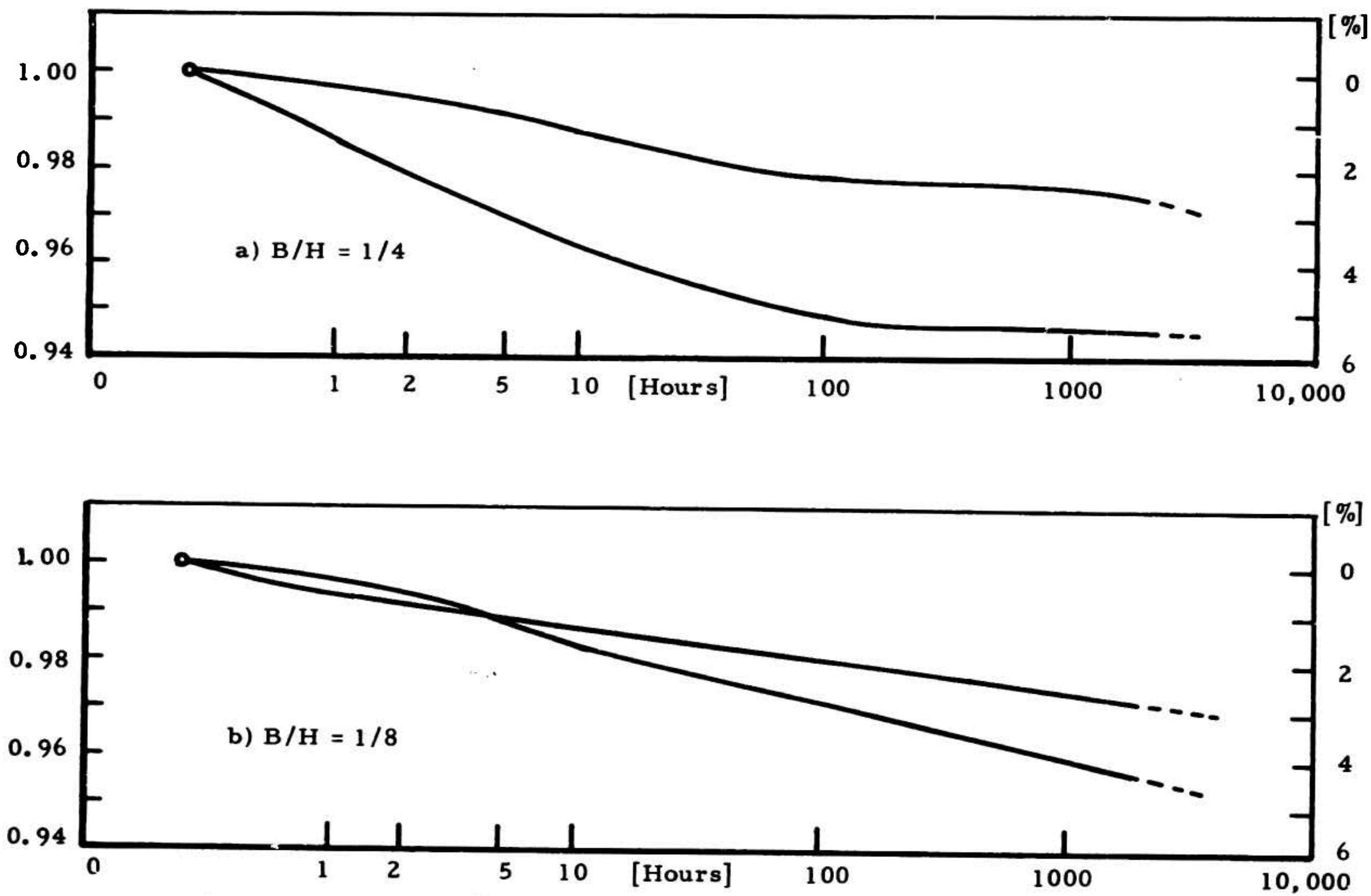

Figure 6. Air Aging at 200°C. Comparison of Best and Worst Case Normalized Curves.
a) B/H = 1/4 b) B/H = 1/8

temperatures, the picture is quite confusing. It seems that aging is worse at 200°C than at 250°C under otherwise comparable conditions. However, closer examination of the shape of the characteristic aging curves for different temperatures during the initial phases of exposure reveals an interesting point: It appears that samples with the higher coercive forces values exhibit a greater loss due to insufficient temperature stabilization level during the first 15-minute period of exposure. Therefore, the approach to a stable "plateau" is a long, continuing process and may take several hundred hours at the lower temperature for these non-prestabilized magnets. Further references will be made to those aging characteristics in the paragraphs comparing best and worst case examples.

d) Air Aging Tests at 150°C

The final elevated-temperature air aging evaluation for samples in the as-received condition was performed at a level of 150°C. These tests were also conducted on samples having a $B/H \approx 1/8$, 1/4 and 1. The results of this evaluation are presented in Tables 10 and 11. Again, a sample (D-1) furnished by the Electron Energy Corporation was included.

As expected, the initial loss after the first 15 minutes of exposure is generally much less than that experienced at the higher temperatures. Subsequent aging of the $B/H \approx 1$ samples for approximately 4000 hours at temperature resulted in additional losses between 0.8 and 2.5%. Aging the thinner samples for only 2000 hours gave increased losses of 1 to 3% for the samples of B/H= 1/4, and 2.7 to 4.6% for B/H = 1/8. While these samples show a greater stability during the total elapsed time than those exposed to higher temperatures, it is quite possible that they have not even approached their plateau. It may be expected that prestabilization at a level higher than the exposure temperature would thermally knock the OCRI down to a level corresponding to that reached at 150°C only after thousands of hours, resulting in improved aging stability at the operating level. Further tests to examine the effectiveness of this procedure were performed. They will be discussed in subsequent paragraphs dealing with attempts at prestabilization.

TABLE 10

ACCELERATED AGING IN AIR AT 150°C, B/H ≈ 1

Sample Number	Initial Loss* (%)	Total Additional Loss Loss** (%)	Total Additional Loss Time (Hours)	Time to Additional Loss of 1% (Hours)	Time to Additional Loss of 3% (Hours)	Initial Properties $_MH_c$ (kOe)	Initial Properties H_k (kOe)	Lot Number
A-17	1.5	0.8	3989	----	----	22.9	7.9	A-1
A-69	1.7	1.7	3989	400	----	37.1	5.60	A-2
A-61	1.6	1.2	3989	1500	----	47.7	9.39	A-2
B-15	1.8	2.5	3989	54	----	37.2	4.69	B-1
B-53	3.8	1.8	3989	56	----	31.6	5.41	B-2
B-52	3.0	1.5	3989	720	----	40 0	6.07	B-2
D-1	1.4	0.5	3473	----	----	26.5	9.7	---

* Drop during first 15 minutes, expressed in % of initial value.
** Drop from the value at 15 minutes, expressed in % of this value.

TABLE 11

ACCELERATED AGING IN AIR AT 150°C,

B/H = 1/8, 1/4

(AS RECEIVED LOT A-2)

Sample Number	Initial Loss* (%)	Total Additional Loss Loss** (%)	Time (Hours)	Time to Loss Of 1% (Hours)	3% (Hours)	Initial Prop. M H_c (kOe)	H_k (kOe)	B/H
A-201	10.9	1.0	1935	1935	--	24.90	4.08	1/4
A-202	6.6	2.9	"	8	--	25.20	3.96	1/4
A-203	8.9	1.5	"	50	--	27.39	4.32	1/4
A-212	7.8	1.7	"	40	--	26.31	5.04	1/4
A-403	6.7	4.1	"	12.5	540	26.40	4.74	1/8
A-410	7.6	3.7	"	36	760	23.70	3.39	1/8
A-408	5.5	2.7	"	14	--	27.50	4.74	1/8
A-411	5.6	4.6	"	11	470	26.10	4.56	1/8

* Drop during first 15 minutes, expressed in % of initial value

** Drop from the value at 15 minutes, expressed in % of this value

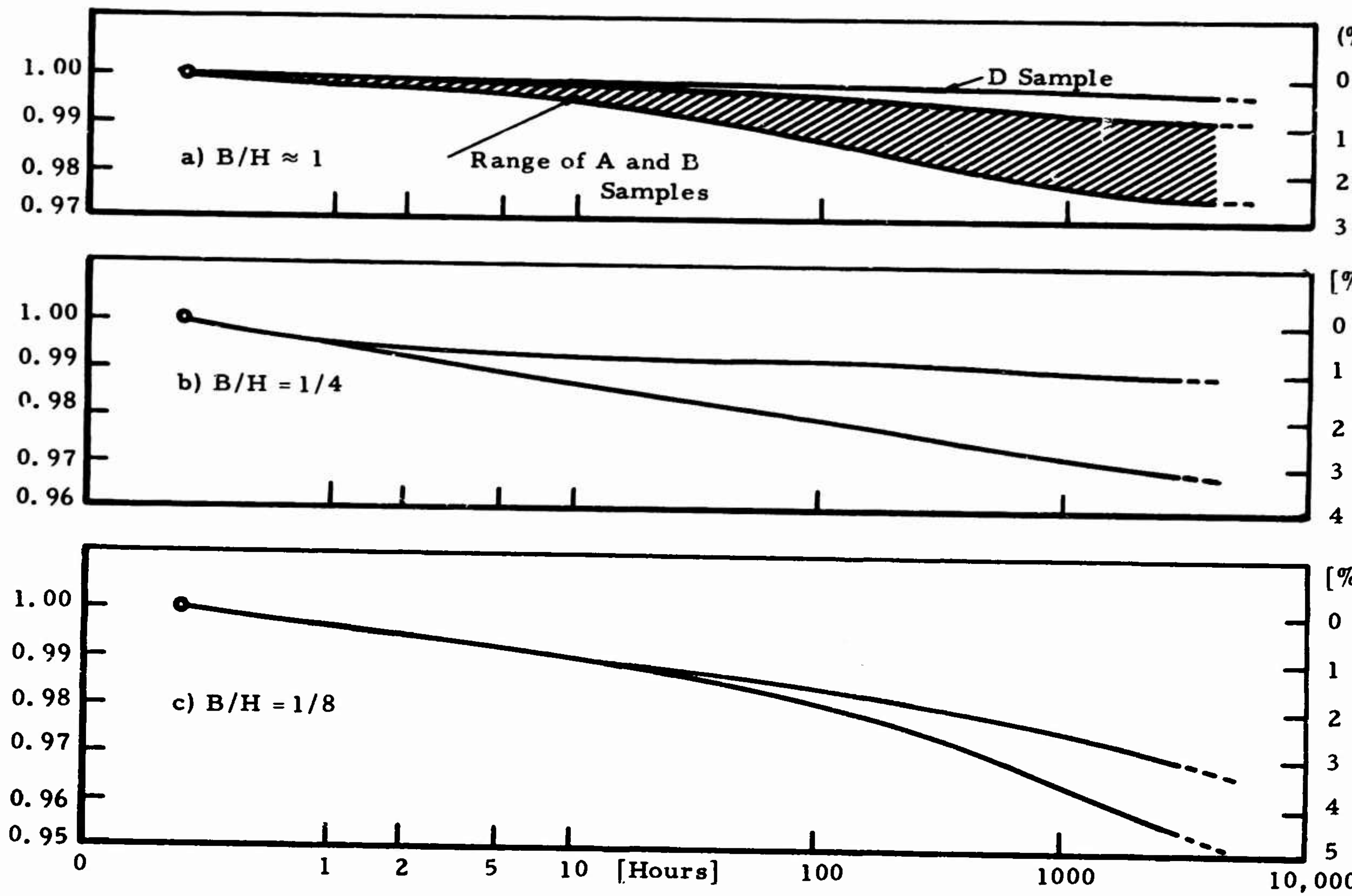

Figure 7. Air Aging at 150°C. Comparison of Best and Worst Case Normalized Curves.
a) B/H ≈ 1 b) B/H = 1/4 c) B/H = 1/8

3. GENERALIZED COMPARISON OF ACCELERATED AIR-AGING CHARACTERISTICS

In a previous report[2] we had described how the observed characteristic aging curves at the various elevated temperatures followed general patterns which could be broken down into 3 phases of interest to the magnet user. The general trends observed then still hold true now after much more testing. Therefore, to aid the reader, we shall repeat the general comments describing these "phases of aging" as an aid to interpreting the figures shown before and the selected curves sketched in Figure 8.

There are very large differences in the aging behavior at any given temperature between the individual magnets, and these do not seem to reflect consistently the differences in origin (and therefore the manufacturing process) or in the initially measured magnetic properties. It is thus not possible to select a set of truly "typical" aging curves. In Figures 2 through 7, we showed curves representing the best and worst aging behavior, respectively, that was encountered in this investigation. These extreme curves were selected from the A and B groups without regard to the origin of the magnets. "Best and worst" were usually judged by the length of the time to an additional loss of 1%. All curves were normalized by calling the flux value after the first 15 minutes unity, and the normalized curves are superimposed at this 15-minute point. We have also shown for comparison additional curves for the Electron Energy Corporation samples where they were included in the tests.

The "instantaneous" irreversible loss incurred during the first interval of heating (15 minutes) is generally continued as a time-dependent "additional aging loss" at a rapidly decreasing rate. These two types are indistinguishable. We refer to the period during which they occur as the first phase of aging, which ends after about 2 hours at $300^{\circ}C$. This is followed by a second phase of a very slow but continuous further reduction of the OCRI which, from the practical point of view, may be considered a plateau with an ill-defined

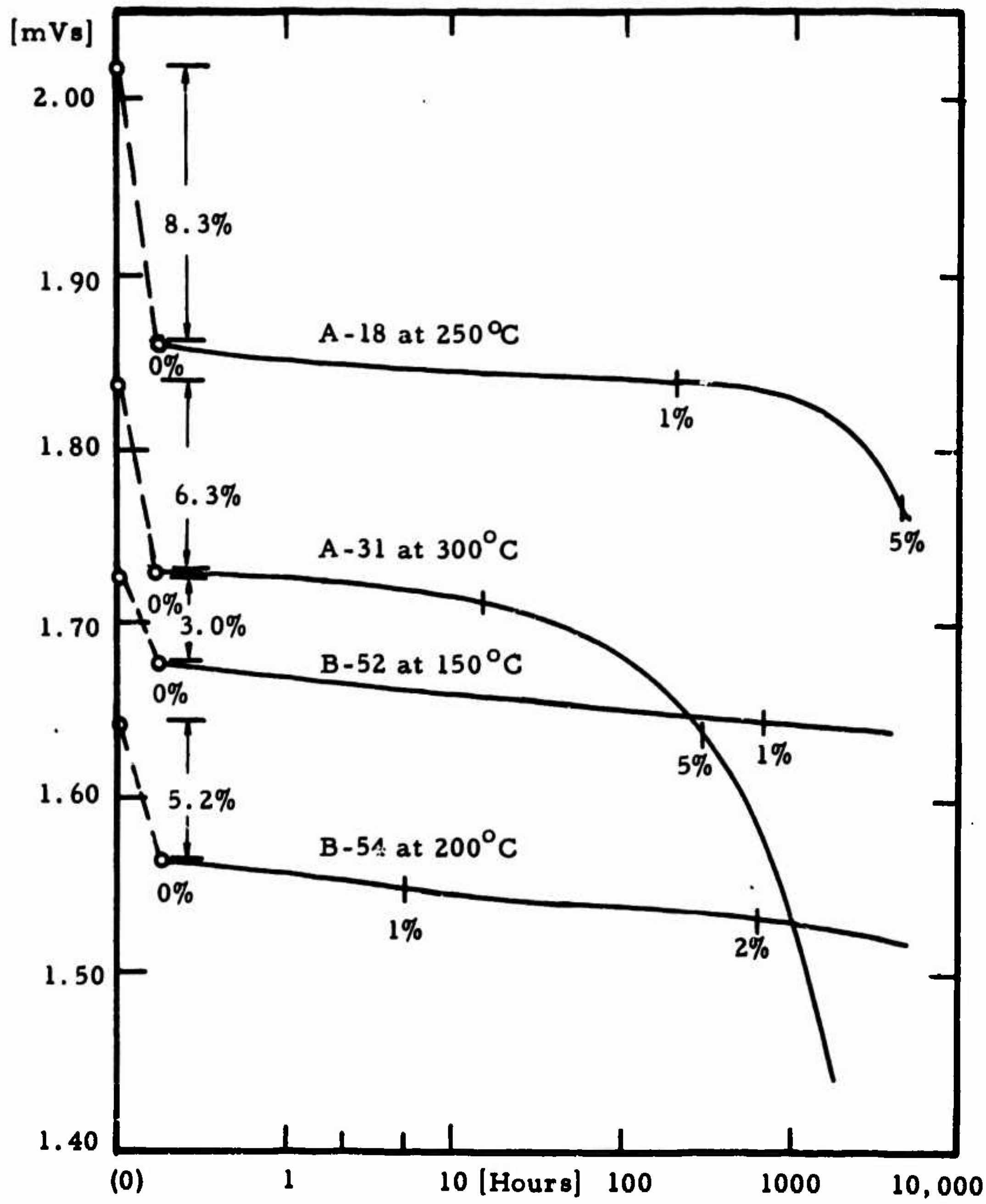

Figure 8. Open Circuit Flux Aging Curve of Four Magnets at Different Temperatures, B/H ≈ 1

termination point depending on the additional loss figure tolerable in a particular application. However, at least at temperatures of 250°C or higher, the rate of decline later accelerates again and the magnet enters a third phase of catastrophic aging during which it becomes eventually useless. As was said before, the differentiation between the "stable" phase 2 and the "catastrophic" phase 3 is rather arbitrary for most - but not for all - magnets studied. The catastrophic decline must, of course, eventually end and the remanence must stabilize again, but perhaps only at a near-zero value. (Note, however, that a few samples entered a second "plateau" of relatively good flux stability after losing only a few percent of their flux, although it had first seemed that the catastrophic phase had already set in.)

The aging curves at 300° and 250°C clearly demonstrate all of the first three stages of aging, and a comparison between them shows that each phase is prolonged as the temperature is lowered. At 200° and 150°C the catastrophic stage had not yet been reached in the tests; in fact, all 150° and all but two - 200°C samples may still have been in the first stage of aging even after the longest exposure times to date.

In paragraph 4 of this section we will further discuss and compare these typical results with those obtained after specific prestabilization heat treatments. Such treatments are aimed at anticipating the entire first aging stage and thus minimizing the additional protracted irreversible loss of a magnet when it is subsequently subjected to long-term thermal exposure.

4. ELEVATED TEMPERATURE AGING AFTER THERMAL PRESTABILIZATION

a) Experimental Procedure

In Section II, paragraph 1, we discussed and illustrated how a substantial improvement in the long-term natural air aging stability of $SmCo_5$ magnets can be derived from specific pre-aging thermal knockdown treatment. We have also indicated that one must also be willing to trade off some percentage of the useful remanence for better long-term stability. For some treatment levels the loss incurred was only a few percent, but at higher levels of treatment, the initial loss incurred was quite significant.

Good stability of magnets at any operating level - room temperature or elevated - , in the sense that the initial time rate of flux loss is minimized, should be achieveable by anticipating the entire first phase of aging and beginning their use in the device just with the onset of the plateau. The previously measured aging curves tell us that this is accomplished in 1/4 to 2 hours at 300° and 250°C. At these temperatures, it would therefore be practicable simply to pre-age at the temperature of future use. But for lower operating temperatures, when the initial aging phase becomes excessively prolonged, a pre-aging by heating to a higher temperature is the only practical solution. It must be recognized that either method will result in a significant reduction of the useful remanence by the thermal prestabilization.

To observe the aging performance of prestabilized magnets we performed the following tests. We arbitrarily chose a temperature 50°C higher than the intended test temperature at which each group of test magnets was pre-aged for 2 hours. The OCRI was then remeasured at room temperature, and each group exposed to its assigned test temperature of 150°, 200° or 250°C.

b) Results of Prestabilization Procedures

The results of these prestabilization treatments are presented in Table 12, and graphically illustrated in Figure 9. In comparing the first four prestabilized magnets (200°C) exposed to 150°C (A-57 to B-46) in this table with data presented in Table 10, a significant improvement in aging stability is now observed. Even the total range bounded by the best and worst-case normalized curves is substantially narrowed. The same observations hold true for magnets prestabilized at 250°C and aged at 200°C.

Again, we observe for the 150°C aging curves that the flux is always in a state of gradual, though small, change with time. This would indicate that, although the 200°C pre-aging heat treatment significantly improved the performance over a magnet exposed in the as-received condition, it was not sufficient for a high degree of stability over several thousand hours. A higher pre-aging temperature, such as 225°C or even 250°C or substantially

TABLE 12

ACCELERATED AIR AGING IN AIR AFTER PRESTABILIZATION *)

B/H = 1

Sample Number	Prestabilization Loss * (%)	Evaluation Temp. (^{o}C)	Total Additional Loss ** (%)	Total Additional Loss Time (Hours)	Time to Additional Loss 1% ** (Hours)	Time to Additional Loss 3% ** (Hours)	Initial Properties $_{M}H_{c}$ (kOe)	Initial Properties H_k (kOe)
A-57	4.5	150	0.35	3046	---	---	39.7	8.0
A-63	2.8	150	0.20	3046	---	---	45.3	10.2
B-68	8.0	150	0.34	3046	---	---	28.0	5.2
B-46	4.8	150	0.38	3046	---	---	38.6	7.4
A-52	7.3	200	0.59	3046	---	---	39.1	7.9
A-48	4.3	200	0.71	3046	---	---	46.9	12.1
B-62	10.2	200	0.72	3046	---	---	28.2	5.8
B-34	11.0	200	0.74	3046	---	---	38.8	6.4
A-77	6.2	250	2.10	3046	640	---	34.4	8.2
A-94	5.5	250	3.51	3046	340	2030	50.0	10.1
B-70	13.5	250	2.57	3046	680	---	30.1	6.0
B-78	7.2	250	2.27	3046	700	---	42.4	7.1

* Prestabilization level 50^{o}C above evaluation temperature.
** Drop from the value after prestabilization treatment, expressed in % of this value.
*** All samples lot-2.

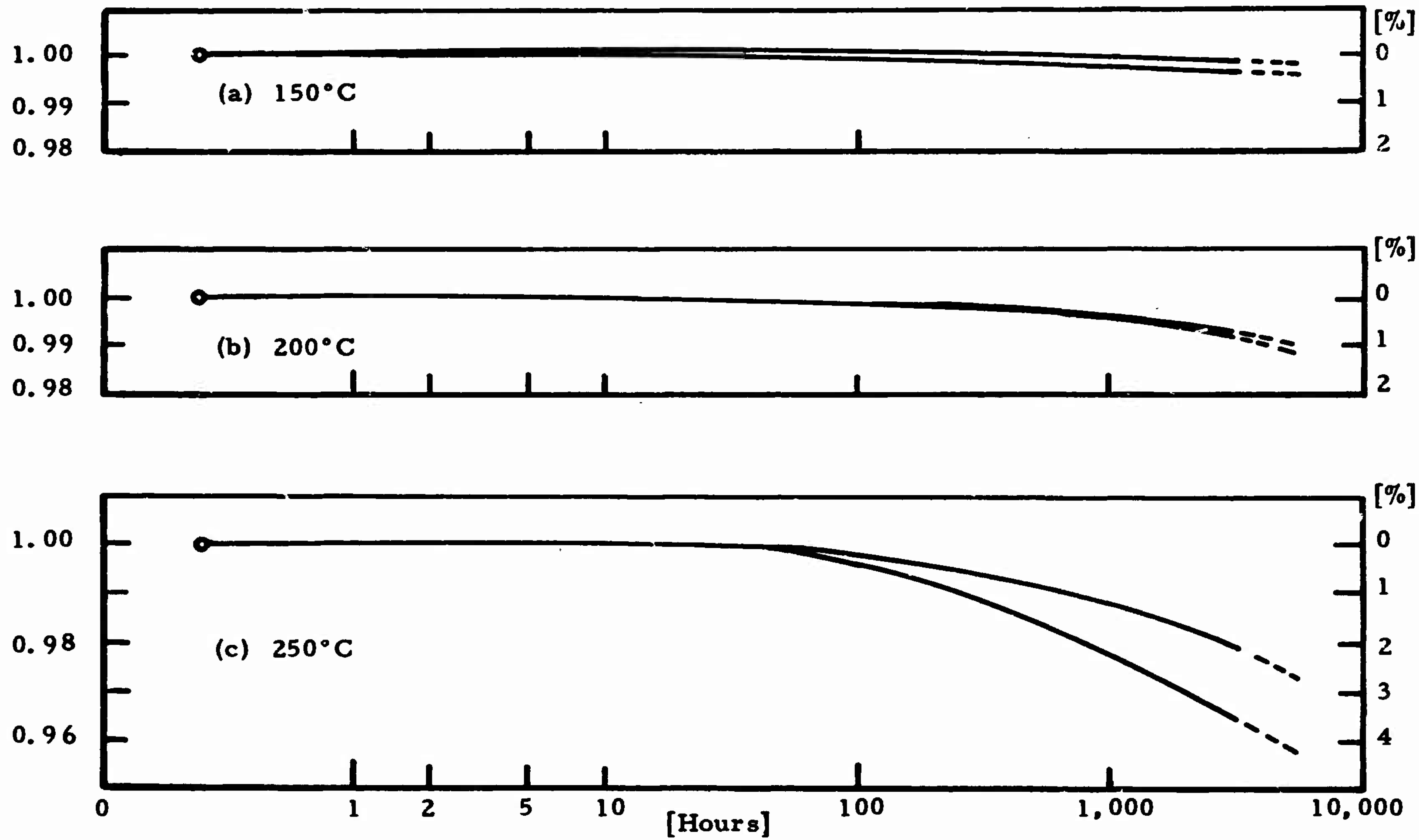

Figure 9. Long Term Air Aging After Prestabilization, $B/H \approx 1$.
(a) 150°C (b) 200°C (c) 250°C

longer preheating at 200°, appears to be necessary to adequately accomplish this.

Our test data also indicates the stability performance of the magnets exposed at 200°C to be slightly better for the short term (~600 hours) than that of the 150° C test magnets. This would indicate the possibility of manipulating pre-aging heat treatments to achieve a different stability performance characteristics for low temperature level applications. That is, to achieve either short-term high stability, or long-term moderate stability.

The test magnets pre-stabilized at 300°C and subsequently aged at 250°C were observed to be improved in the sense that, for the best case, the short term stability is better than without pre-stabilization up to ~700 hours. From that point on, the rate of loss increases gradually, but we see no evidence at the end of ~3000 hours of approaching a truly catastrophic loss phase like that which was seen in Figure 3 for the "as received" samples aged at 250°C. For a given prestabilization temperature, the loss is roughly inversely proportional to H_k. For a given H_k value, the flux reduction during preheating at 250° and at 300°C is about the same, but it is less - only about 1/2 to 2/3 - at 200°. This indicates that thermal wall relaxation is complete after 2 hours above 250°C, while 2 hours at 200° may not be sufficient to achieve a magnetic equilibrium state. The previously mentioned higher rate of initial flux change during subsequent aging at 150°C is consistent with this view.

The relationship of the prestabilization loss with ${}_MH_c$ is qualitatively the same, but less systematic in detail than the correlation with H_k.

By contrast, the additional aging losses cannot be correlated with H_k or ${}_MH_c$. In fact, they are fairly constant for a given prestabilizing and aging test temperature and independent of H_k or H_c for 200°/150° and 250°/200°. These losses increase, however, with increasing temperature; quite drastically so - by a factor 3 to 5 - between 250°/200° and 300°/250°. This indicates rather clearly that the long-term aging losses are metallurgical in orgin and not primarily changes in the domain pattern as are the prestabilization losses. The unusually high additional loss for the high - ${}_MH_c/H_k$ sample A-94 is thought to be caused by structural factors, perhaps a higher degree of porosity.

Generally speaking we can say that the flux stability during the first several hundred to several thousand hours of operation at an elevated temperature can definitely be improved by a thermal pre-aging treatment. If a designer can specify the magnet performance desired in a defined thermal environment, then either he or the producers should be able to prescribe the proper pre-aging treatment required. The present results can serve as a basis for this. However, additional experiments with larger numbers of current-production magnets should yield more complete sets of empirically derived thermal treatment data for various special applications.

5. EFFECTS OF THERMAL AGING ON THE DEMAGNETIZATION CURVE

In an attempt to learn a little more about the causes of the loss of the open-circuit remanence in each evaluation than can be gleaned from measurements of the OCRI flux alone, we have traced a demagnetization curve on the samples before the start and after the termination of the thermal exposure test.

Previously we had presented a detailed analysis of characteristic intrinsic and normal demagnetization curves for a few specific samples which were exposed at the higher temperatures and terminated after sustaining severe losses of open-circuit remanent induction.[2] Due to the large number of samples with different L/D ratios and treatments, evaluated at various temperatures to tabulate every parameter with loss differences after aging (before and after recharging) and to relate these to the initial parameter values would be a large undertaking. Since a study of the causes of aging was not a principal objective of the work under this contract, such a detailed critical analysis of the data is unwarranted. However, to utilize the material that has been recorded, we surveyed the sets of demagnetization curves for each specific evaluation temperature to determine the extreme cases as judged by the changes observed in intrinsic coercivity and residual induction.

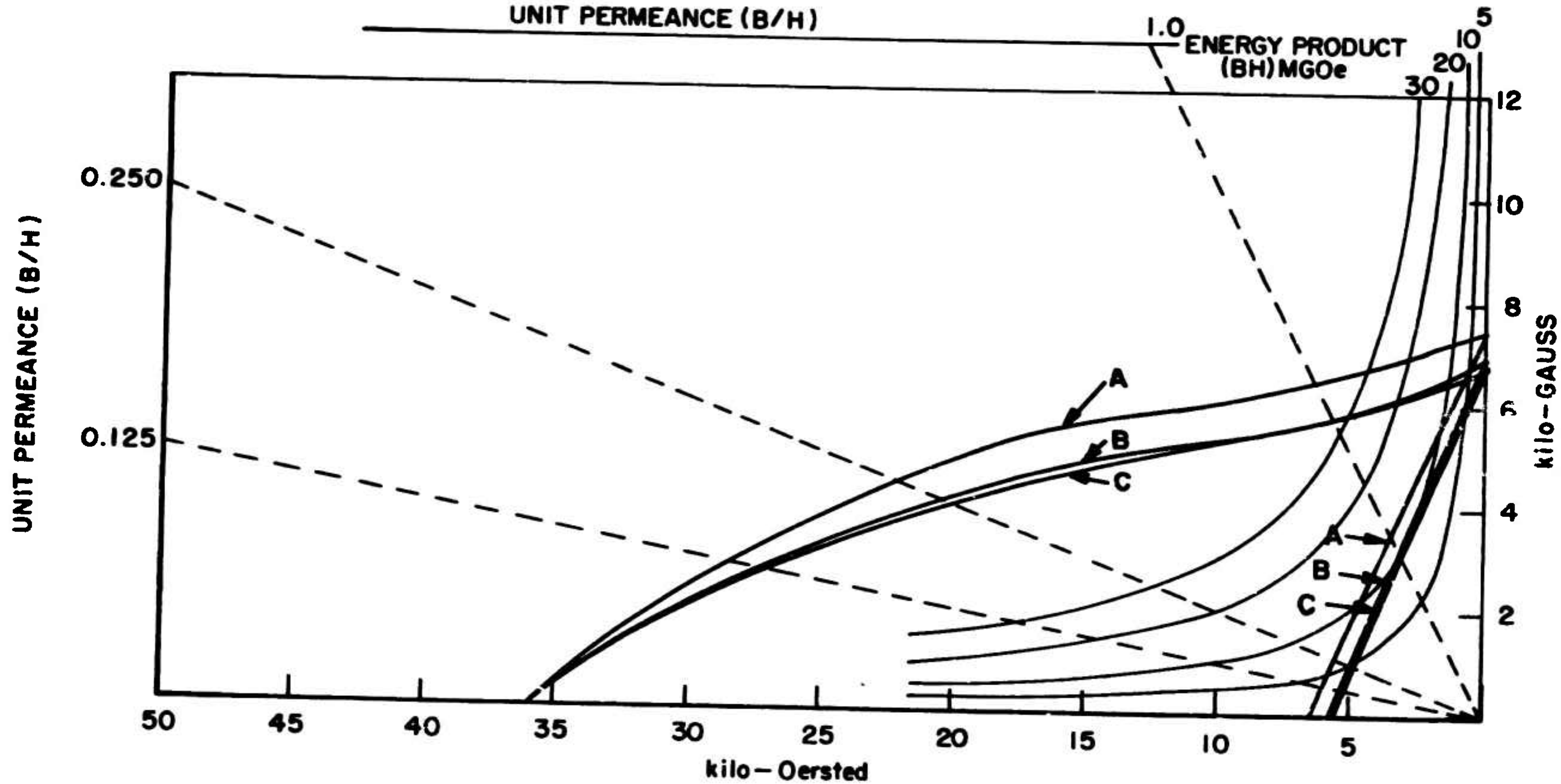

Figure 10. Thermal Aging Effects on Demagnetization Curve. Sample No. B-15.
A. Before Aging B. After 4,000 hours at 150°C in Air,
C. Remagnetized

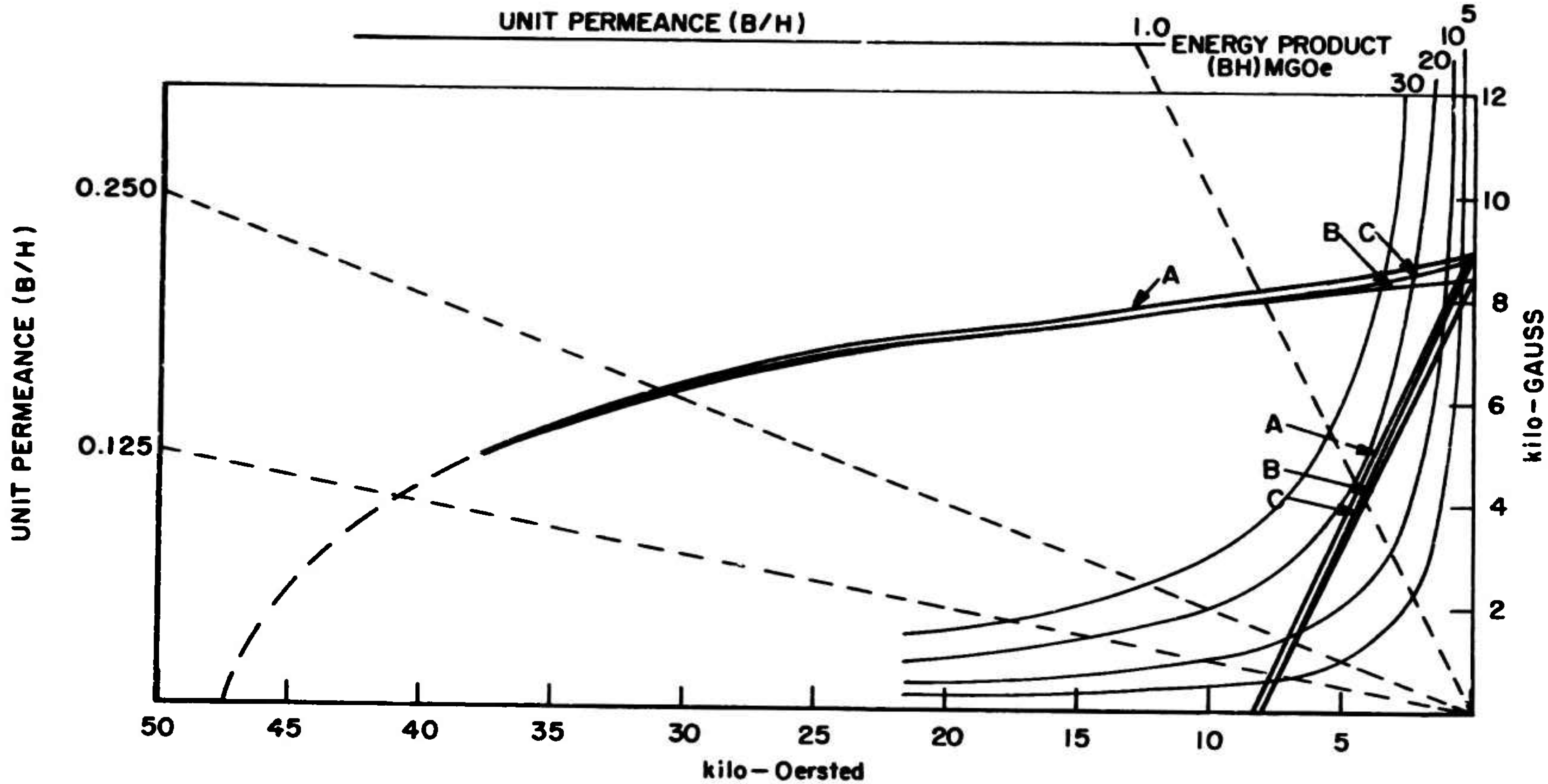

Figure 11. Thermal Aging Effects on Demagnetization Curve. Sample No. A-61.
A. Before Aging B. After 4,000 hours at 150°C in Air,
C. Remagnetized

For the Figures 10 through 18, we have selected (B-H) vs. H and B vs. H room-temperature demagnetization curves of 9 samples which are representative of the range of properties observed before and after the aging tests from 150° to 300°C. The observed change (or lack of change) during the thermal exposure is graphically shown, and one can also judge the extent to which losses are recoverable by recharging.

Each sample then has three sets of curves: one (A) measured bef. . the start of the exposure; a second one (B) determined right after the termination of heat-aging and traced down from the remanence point by inserting the sample into the measuring yoke and applying an increasing negative field first; and a third set (C) measured after remagnetizing the sample in the original direction (with +34 to +38 kOe, depending on the measurement configuration in the yoke).

Inspection of the curves in Figure 10 indicates that at the lower temperature level of exposure (150°C) there is no noticeable change in ${}_{M}H_{c}$ during aging, even after 4000 hours. For the sample of Figure 11, the ${}_{M}H_{c}$ was greater than the available maximum field. However, the near coincidence of curves A, B and C at high fields suggests that here, too, the coercivity did not change during exposure. The residual induction, B_r, is reduced by 5-10%; the induction coercive force, ${}_{B}H_{c}$, follows B_r. The loss of remanence is largely recoverable by recharging for magnets with high H_k (good loop squareness), such as that of Figure 11, but not when H_k is low (Figure 10).

In Figures 12 and 13, we observe that as the aging temperature level is increased to 200°C, there is now a loss of the intrinsic coercive force loss. This loss is a small portion ($\sim$ 5%) of the total ${}_{M}H_{c}$ value; it is not recoverable upon recharging. A larger fraction (12-15%) of residual induction B_r and the OCRI is lost at this temperature. As before, those magnets with a high H_k number were observed to partially recover these properties after recharging better than those of lower H_k.

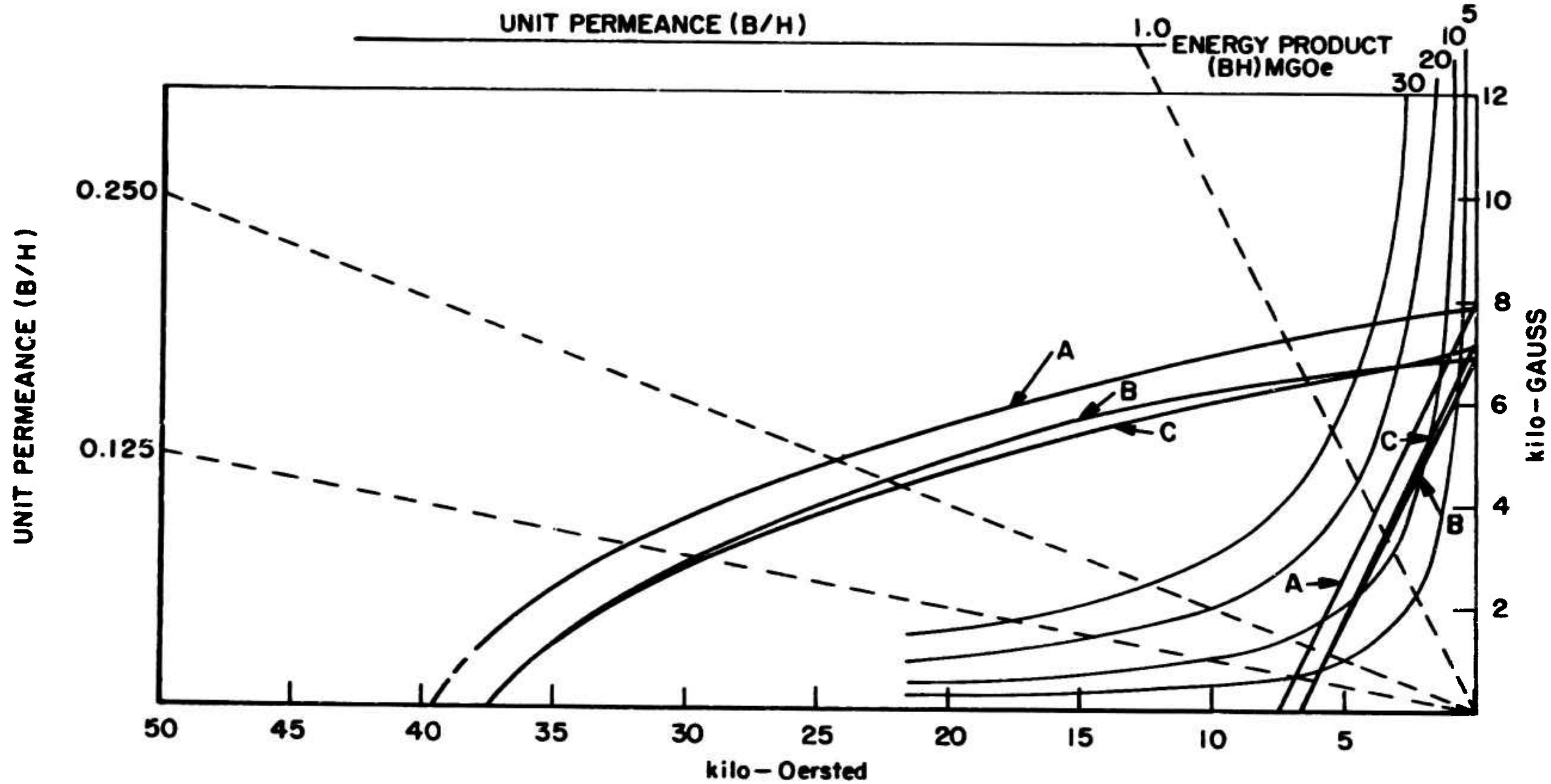

Figure 12. Thermal Aging Effects on Demagnetization Curve. Sample No. B-48.
A. Before Aging
B. After 4,000 hours at 200°C in Air
C. Remagnetized

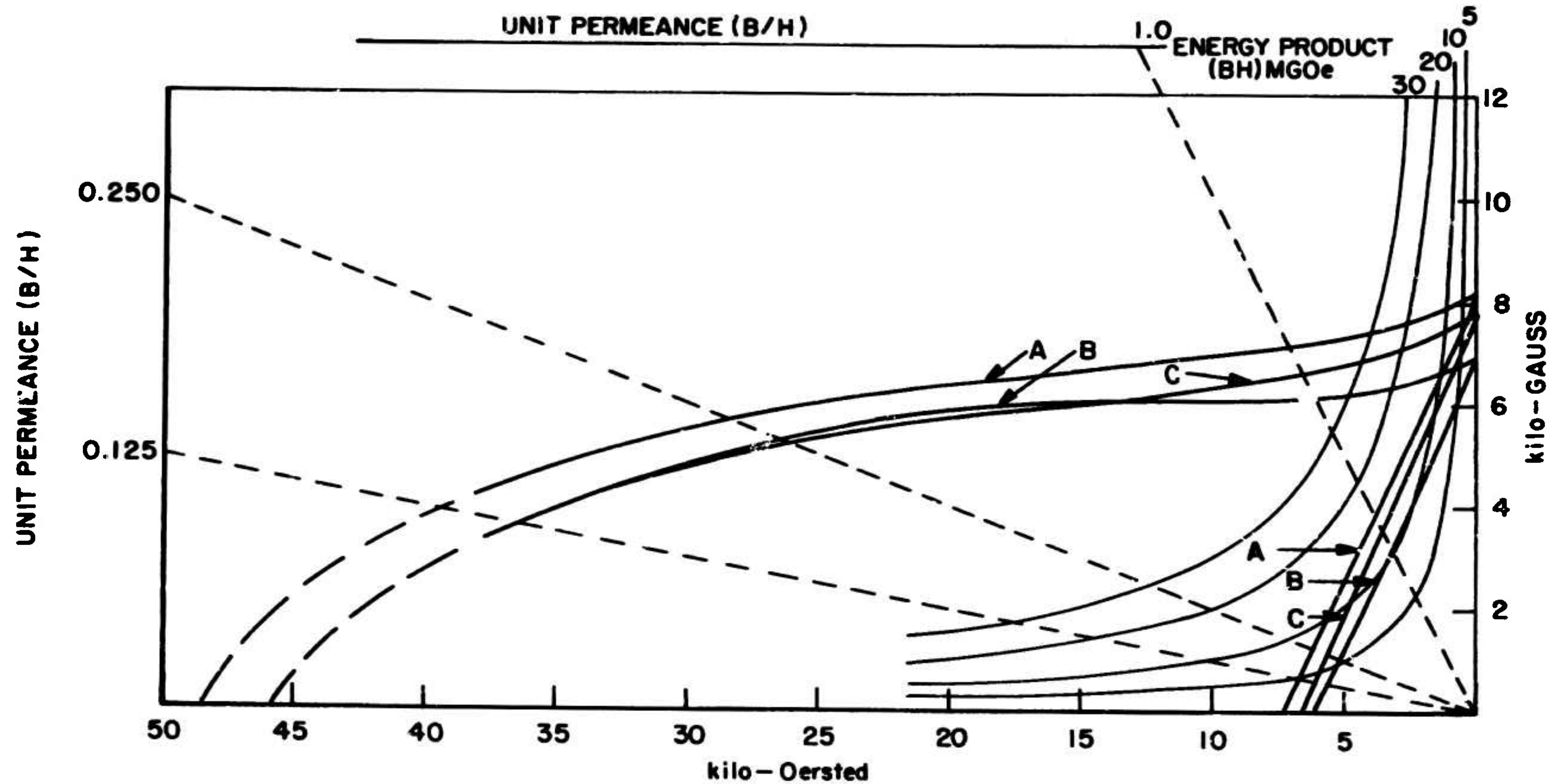

Figure 13. Thermal Aging Effects on Demagnetization Curve. Sample No. A-62.
A. Before Aging B. After 4000 hours at 200°C in Air
C. Remagnetized

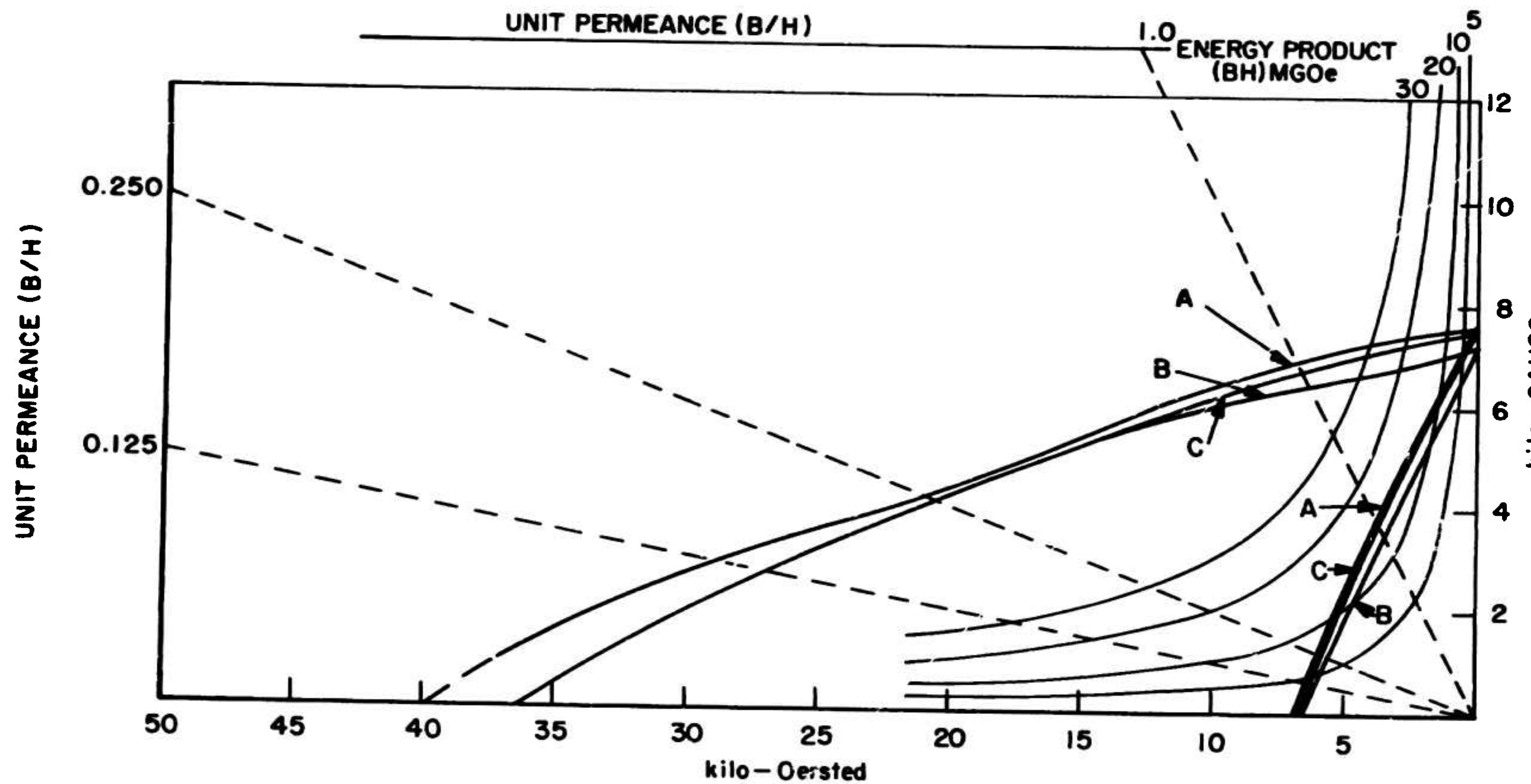

Figure 14. Thermal Aging Effects on Demagnetization Curve. Sample No. B-25.
A. Before Aging B. After 4400 hours at 250°C in Air
C. Remagnetized

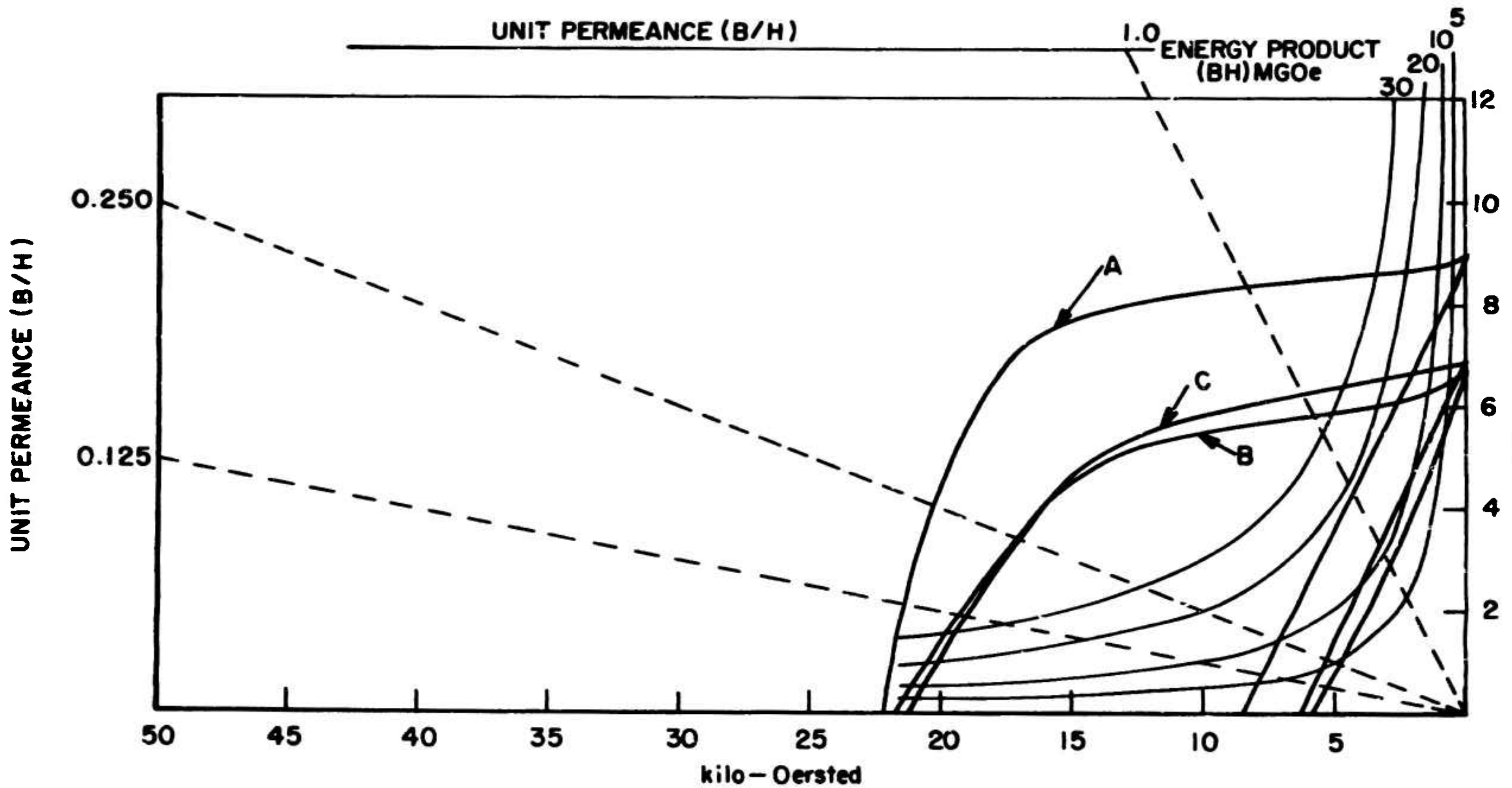

Figure 15. Thermal Aging Effects on Demagnetization Curve. Sample No. A-16.
A. Before Aging B. After 5000 hours at 250°C in Air
C. Remagnetized

At the aging temperature level of 250° C we generally observe the same trends as at 200°C, but quantitatively larger effects. A representative set of curves (Figure 14) shows an increased fractional loss 5-10%) of intrinsic coercive force. This loss was sustained by most magnets and was generally not recoverable on recharging. The loss of residual induction and its recoverability on recharging were again found to be highly dependent on the magnitude of the intrinsic coercive force and the squareness of the demagnetization curve. Figure 15 shows an example of aging effects on a magnet with a relatively square demagnetization curve ($H_k \approx 12$ kOe), but a low intrinsic coercive force. A severe loss of residual induction was incurred during the initial 15 minutes of aging. Subsequent aging reduced B_r and OCRI even further. Recharging of this magnet resulted in a slight decrease in $_MH_c$, while the knee of the curve became more pronounced again. The loss of B_r was essentially unrecoverable.

At the highest temperature level studied (300°C), we observed a wide variation of aging characteristics. The Figures 16 through 18 illustrated this with three sets of curves which are representative of the range of aging characteristics observed and demagnetization curves encountered.

In Figure 16, we observe the same general magnetic-property/aging-loss pattern as experienced with a magnet having a low coercive force and square hysteresis loop at 250°C (see Fig. 15). However, upon recharging we observe the surprising fact that, while the intrinsic coercive force is higher than after aging, a lower remanence is incurred. The shape of the curve in the low negative field region would strongly indicate the presence of a second soft-magnetic metallurgical phase in the magnet.

Magnets whose typical demagnetization curves are best represented by those shown in Figure 17 (with high intrinsic coercive force, but no semblance of a knee), are observed to experience an almost parallel translation toward lower value of the second-quadrant curve upon aging. After recharging these magnets, the slope of the aged intrinsic curve increases (that is, B_r increases), but the coercivity remains fixed at the $_MH_c$ value

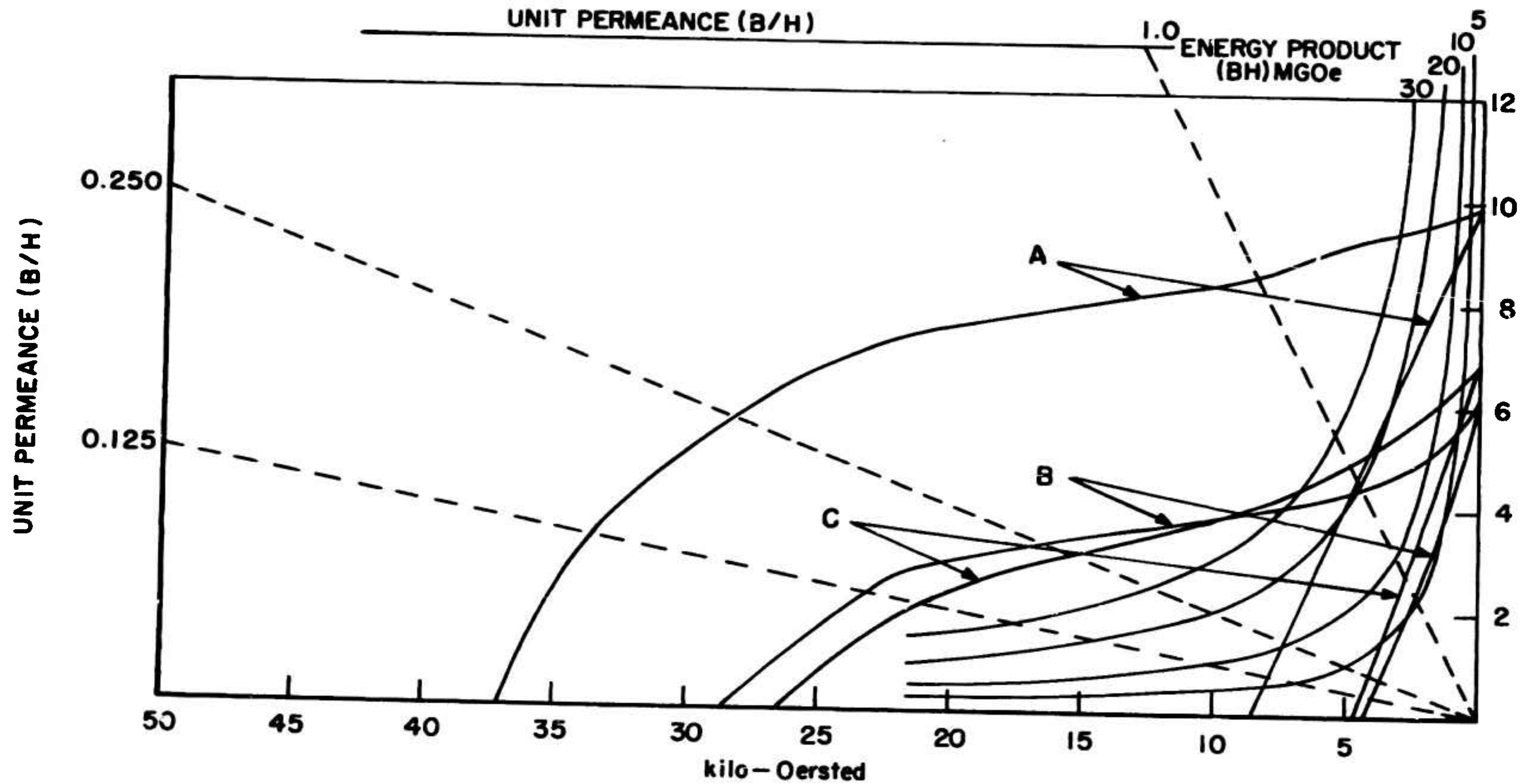

Figure 16. Thermal Aging Effects on Demagnetization Curve. Sample No. A-40.
A. Before Aging B. After 1250 hours at 300°C in Air
C. Remagnetized

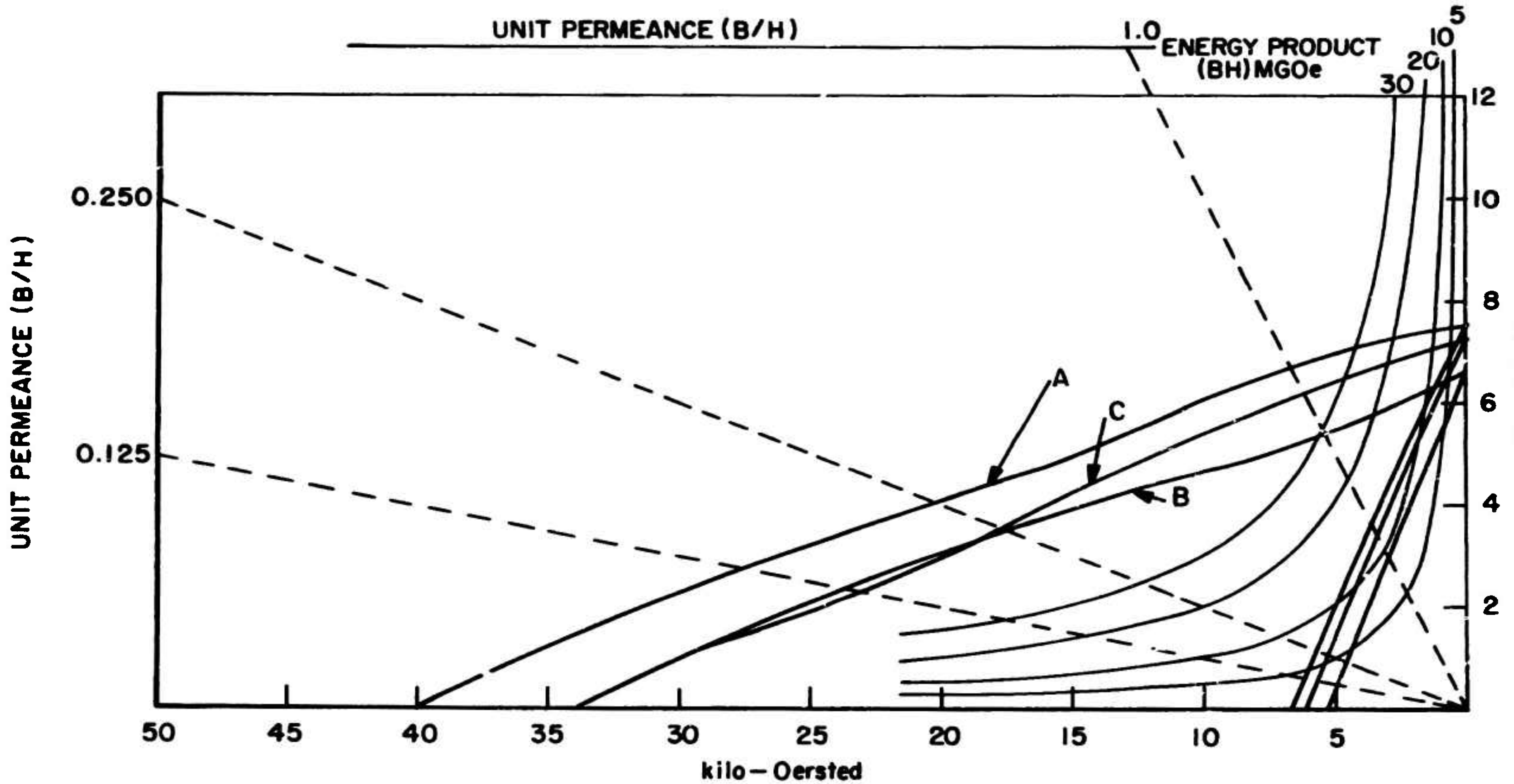

Figure 17. Thermal Aging Effects onDemagnetization Curve. Sample No. B-31.
A. Before Aging
B. After 2255 hours at 300°C in Air
C. Remagnetized

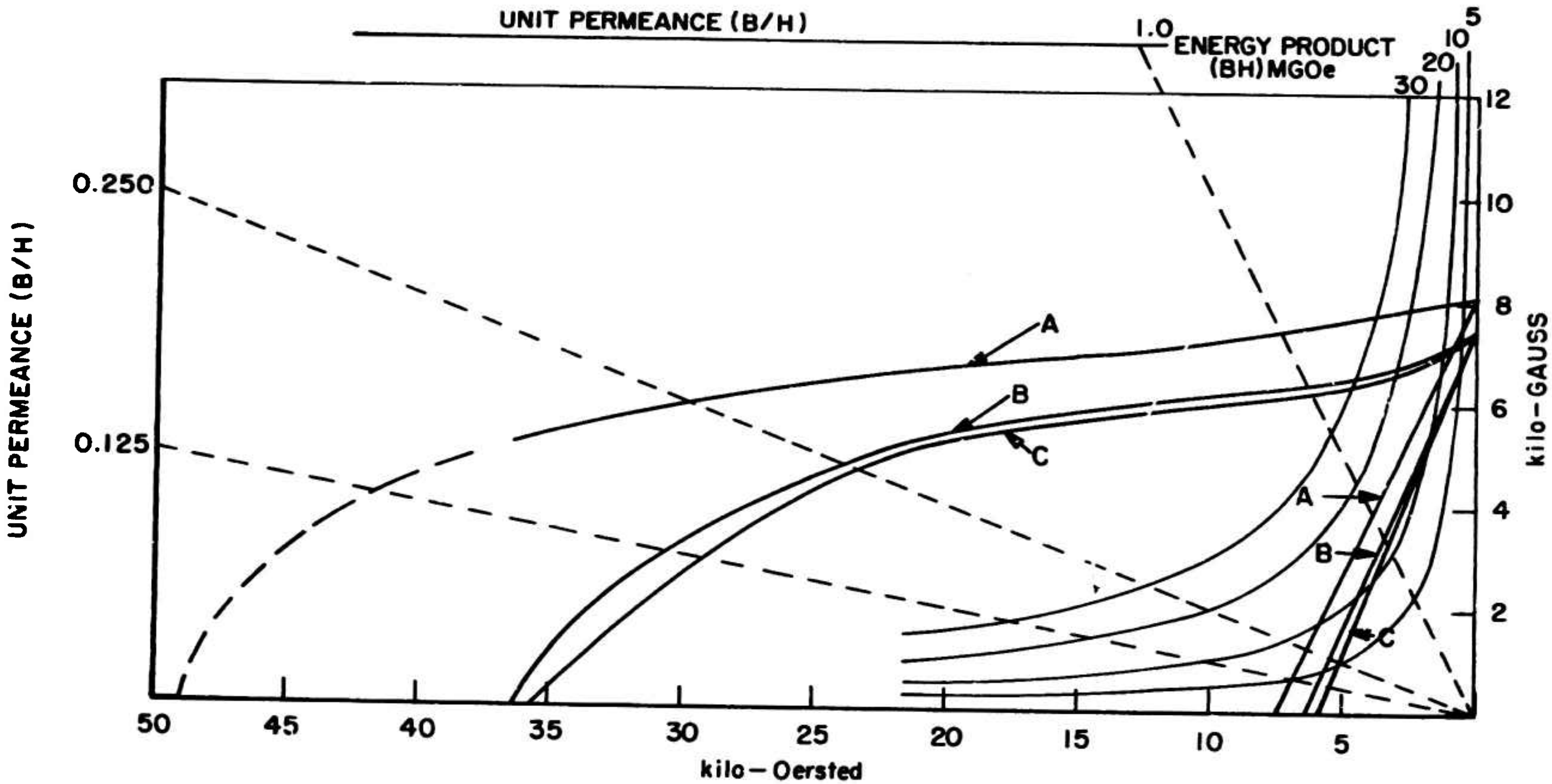

Figure 18. Thermal Aging Effects on Demagnetization Curve. Sample No. A-25.
A. Before Aging B. After 3615 hours at 300°C in Air
C. Remagnetized

reached after aging.

For magnets with high ${}_MH_c$ values and a generally square loop, as shown in Figure 18, we observe that a large loss in ${}_MH_c$ is sustained during aging at 300°C. The residual induction value B_r decreases only by a small fraction of the original value on long-term heating. The curve shape becomes distorted and shows a dip at low negative applied fields after aging and after recharging. This may be due to a second soft-magnetic phase developing in the material at this temperature. Upon recharging, the B_r value increases only a minimal amount. But, the curve shapes are now somewhat different. The curves B and C cross over as ${}_MH_c$ is lowered as B_r increases during recharging.

From these observations it generally appears that both the degree of property value losses and general shifting of the curves is not only dependent on the temperature level of exposure, but also on the shape of the curves before the elevated-temperature exposure. At the lower exposure-temperature levels of approximately 100° to 200°C, little or no change in ${}_MH_c$ is observed. What little coercivity loss is incurred during aging appears to be permanently fixed and can not be recovered by recharging. The residual B_r may be partially or almost completely recoverable, dependent on the squareness of the loop shape.

At higher aging temperatures of approximately 250° to 300°C, samples with low coercive force and poor to modest loop squareness will sustain less of a loss in ${}_MH_c$ and B_r. Losses of both properties incurred at these temperatures are essentially permanent-recharging does not significantly increase ${}_MH_c$ or B_r over the values at the end of aging.

SECTION III

THERMAL CYCLING TESTS ON $SmCo_5$ MAGNETS

1. EXPERIMENTAL PROCEDURES

This section deals with measurements of the changes of the open-circuit remanent induction flux (OCRI) during thermal cycling of magnets between room temperature and the extremes of +300°C and -100°C. The pull-coil apparatus used for these tests was described in the first report.[1] The temperature coefficients of samples 1/4" diameter and L/D = 0.4 ($B_d/H_d \approx 1$), were presented in the second report.[2]

We now have completed measurements on samples with a geometry yielding B/H unit permeance values of B/H = 1/8, 1/4, and 2-1/2. Some of these measurements have been performed on samples in the as-received condition, and others - after machining to size from larger stock magnets. All test magnets were 1/4" in diameter. The measurement data presented in Table 13 lists the individual test magnet thermal coefficients and previous treatment. Specific machining treatment will be elaborated on in Section IV. The average value of specific permeance figures for each manufacture is presented in Table 14.

The general procedure was to heat or cool the samples in a step-wise fashion, taking measurements of the OCRI at approximately 25° intervals when the temperature had nearly, but not completely, stabilized. The aim was to obtain a close correspondence between remanence and temperature readings while keeping the cycle time and exposure to high temperatures to a minimum. Typical time periods required for the different phases of the measuring cycles were: 15-20 minutes for heating from room temperature (RT) to +250°C or 300°C: 1 hour to cool back to RT.

After it had been verified that the exposure-time element in these tests was negligible, we routinely prestabilized the magnets by heating them to 250°C for 2 hours, giving them a "knockdown" similar to that routinely applied by the manufacturers of magnets for traveling-wave-tube use. A typical experimental sequence for these samples after prestabilization was as follows:

$$RT \leftrightarrow 250^{o}C \ldots 2 \text{ cycles}$$
$$RT \leftrightarrow 300^{o}C \ldots 2 \text{ cycles}$$

The samples used in these test were again selected from both, the A and B lots, such that the upper and lower extremes of ${}_{M}H_{c}$ and H_k were represented. We have also included data based on one sample of each configuration from the Electron Energy Corporation.

From examining the plots of OCRI versus temperature for the samples identified in Table 13 we observed that none of the curves were straight lines. They reflect the general shape of the temperature dependence of the saturation magnetization.

2. NUMERICAL VALUES OF THE REVERSIBLE TEMPERATURE COEFFICIENTS

We calculated a number of temperature coefficients of the open-circuit flux change from the reversibly traced curves of each magnet configuration between 25° and +300°C. Listed in Table 13, are the values of the slope at the fixed temperatures +25° C and +300°C. Also given are mean reversible thermal coefficients for the temperature ranges +25°C to 250°C, and +25°C to +300°C as determined from the coordinates of the two end points of each range. We have also listed the previous history of each sample before the test, that is, whether it was in the "as-received" condition or how it was machined.

In contrast to the aging behavior, very little variation of the reversible temperature coefficients was found among the magnets tested in each configuration. The small differences which were observed do not seem to correlate in any manner to the initial magnetic properties. They should relate to the chemical composition of the magnets, e.g., the purity of the samarium metal used, and also the Sm-to-Co ratio and therefore to the

TABLE 13

THERMAL COEFFICIENTS

$(\Delta B_d/B_d)/\Delta T$ At B_d/H_d = 1/8, 1/4 and 2-1/2

Sample Number	At Specific Points +25°C %/°C	+300°C %/°C	Over Temperature Range°C 25 to 250 %/°C	25 to 300 %/°C	B_d/H_d	General Comments *)
A-407	0.031	0.063	0.036	0.040	1/8	As Received
A-435	0.027	0.064	0.033	0.040	1/8	As Received
A-219	0.039	0.061	0.041	0.044	1/4	As Received
A-226	0.034	0.060	0.040	0.044	1/4	As Received
A-624	0.033	0.059	0.041	0.044	1/4	Ground-Rate 1
A-602	0.036	0.057	0.040	0.043	1/4	Ground-Rate 1
A-623	0.037	0.057	0.042	0.044	1/4	Ground-Rate 1
A-633	0.032	0.060	0.042	0.046	1/4	Ground-Rate 2
A-616	0.036	0.056	0.042	0.044	1/4	Ground-Rate 2
A-620	0.033	0.057	0.040	0.043	1/4	EDM-Rate 4
						EDM-Rate 3
B-215	0.037	0.068	0.042	0.045	1/4	Gr. to Thickness
B-234	0.036	0.068	0.044	0.045	1/4	Gr. to Thickness
A-516	0.038	0.069	0.045	0.047	2-1/2	Ground-Rate 1
A-525	0.038	0.070	0.044	0.046	2-1/2	Ground-Rate 1
A-526	0.037	0.071	0.046	0.047	2-1/2	Ground-Rate 2
A-500	0.037	0.069	0.044	0.047	2-1/2	Ground-Rate 2
A-505	0.039	0.074	0.045	0.048	2-1/2	EDM-Rate 4
A-511	0.038	0.074	0.045	0.048	2-1/2	EDM-Rate 4
A-520	0.040	0.069	0.046	0.047	2-1/2	EDM-Rate 3
A-506	0.037	0.071	0.045	0.047	2-1/2	EDM-Rate 3
B-519	0.044	0.077	0.048	0.052	2-1/2	Ground-Rate 1
B-510	0.044	0.078	0.048	0.051	2-1/2	Ground-Rate 1
B-524	0.040	0.079	0.049	0.051	2-1/2	Ground-Rate 2
B-542	0.041	0.077	0.049	0.051	2-1/2	Ground-Rate 2
B-537	0.040	0.077	0.049	0.051	2-1/2	EDM-Rate 4
B-535	0.040	0.079	0.050	0.052	2-1/2	EDM-Rate 4
B-544	0.040	0.079	0.049	0.052	2-1/2	EDM-Rate 3
B-545	0.040	0.079	0.050	0.052	2-1/2	EDM-Rate 3

*) See Section in Machining Procedure Information.

balance of the metallurgical phases present. Likewise the machining methods and rate of material removal in this evaluation indicated no noticeable effect on coefficients.

The average values of the specific data presented in Table 13 is summarized in Table 14 for the various configurations and source of manufacture. The reversible temperature coefficient values reported for each configuration are generally characteristic for sintered $SmCo_5$ magnets and largely unaffected by differences in origin of the magnets and the manufacturing methods used in producing them. The range of values measured by us agree rather well with numbers published by the magnet producers in their sales literature (even though these often apply to the zero-field residual induction). There is one difference however, in the trend of the reversible temperature coefficient data we have measured and that published by Raytheon.[6] Contra· .. their literature we have observed an increasing coefficient number ‸ function of increasing B/H values over a temperature range of 25 tc 250°C.

6. Raytheon, Technical Bulletin No. PT-3233, February 1972.

TABLE 14. THERMAL COEFFICIENTS AVERAGE VALUES

B_d/H_d = 1/8, 1/4, 1 and 2-1/2

B_d/H_d	Slope at Temperature °C			Over Temperature Range °C			Manufacturer
	-100 (%/°C)	25° (%/°C)	+300 (%/°C)	-100 to +25 (%/°C)	25 to 250 (%/°C)	25 to 300 (%/°C)	
1/8	---	0.029	0.063	---	0.034	0.040	General Electric
1/4	---	0.035	0.058	---	0.041	0.044	General Electric
1	0.031	0.036	0.056	0.032	0.044	0.042	General Electric
2-1/2	---	0.038	0.071	---	0.045	0.047	General Electric
1/4	---	0.036	0.068	---	0.043	0.045	Raytheon
1	0.031	0.039	0.062	0.035	0.045	0.047	Raytheon
2-1/2	---	0.040	0.078	---	0.049	0.051	Raytheon
1/8	---	0.036	0.070	---	0.047	0.048	Electron Energy
1/4	---	0.037	0.067	---	0.046	0.048	Electron Energy
1	---	0.035	0.063	---	0.045	0.048	Electron Energy

SECTION IV

MACHINING EFFECTS ON LONG-TERM AGING PERFORMANCE

1. PURPOSE OF MACHINING INVESTIGATION

In some magnet applications the magnet user may wish to perform minor surface machining operations on stock shapes purchased from a producer. Our purpose then is to determine whether the removal of surface material from magnets in the "as-received" condition by accepted methods have either a beneficial or detrimental effect on their long-term accelerated aging performance.

In this investigation we have prepared samples from stock magnets by three material removal methods, surface grinding, centerless grinding, and electric discharge machining. The grinding methods of surface material removal are similar in a sense to that used in the production of isopressed samples. That is, the pressed bar is surface ground to diameter, and then sliced to thickness with possibly an end surface grinding pass to standard thickness. One would expect relatively little noticeable effect on the aging performance or the magnet properties due to additional machining - provided proper care is exercised in handling the brittle material and a reasonable rate of material removal is used.

On diepressed samples where the finished magnet diameters are anticipated in the pressing-sintering operation, there may be some noticeable effect in reducing sample diameters (or thickness) of larger stock size magnets.

Regardless of the method of production, if there were any misalignment or inhomogeneity of powder in the pressing the green magnet, a change in property may be observable as a result of machining.

2. SAMPLE PREPARATION

Samples machined in this investigation were prepared for two different evaluations. One evaluation called for samples centerless ground and electric discharge machined at two different rates of material removal.

Also, when placing our order for die-pressed test magnets we were informed that it would be inadvisable to produce samples in the finished state with such short configurations (B/H = 1/4 and 1/8) in the diameter we specified. This is mainly attributed to difficulties encountered in maintaining powder alignment and distribution in the die, and a high sample mortality rate. As we wished to work with a minimum of sample diameters, etc., for comparison purposes in the aging studies, we decided to machine these at our own facility. The second machining effort then was directed toward surface grinding Raytheon samples 0.250" diameter, 0.100" thickness down to a thickness of 0.025" and 0.012".

a) Centerless Grinding

Stock GE and Raytheon magnets of 0.500" diameter, 0.200" thickness, and 0.025" thickness (G. E. only), were reduced in diameter to 0.250" to provide samples with a B/H = 2 1/2 and 1/4 respectively. The centerless grinding operation was performed under our supervision at a local facility (Specialty Machines, Inc., Dayton, Ohio), specializing in this service. To conform with their experience in centerless grinding permanent magnets, we pulse demagnetized the magnets in a 60 kOe field as best we could. A composite stack of the thin samples was prepared by bonding them together with a quick drying cellulose cement. All samples to be ground at a given rate (both GE and Raytheon) were aligned in a row and then fed into the grinding machine. In Tables 15 through 20, we have identified the grinding rate of material removal at 1 and 2. These numbers refer to the amount of material removed during each pass of the string of magnets through the operation. Rate 1 was at .001" reduction in radius each pass, and Rate 2 - .002" each pass.

The equipment used was a Cincinnati #2 centerless grinder with an aluminum oxide wheel (80L), 8" long, 20" diameter, rotating at 1150 rpm (~6000 sfpm). The work piece drive wheel rotated at 20 rpm. Frequent adjustments (~ every 10 passes) were made on the wheel spacings and guide bar height for decreasing sample diameter, and the aluminum oxide wheel dressed every other adjustment. A water soluble grinding liquid coolant (Stewart's Supercool) was used during the operation.

Generally speaking the grinding operation went quite well. Only one of the thin samples was cracked in each operation (Rate 1, Rate 2). No evidence of edge chipping could be found on any of the samples. The most critical point in the operation is the first few passes during which "high" spots are removed and the samples become more symetrically round. One noticeable difference between the isopressed samples and diepressed samples was observed during the initial removal of ~ 0.010" of material. Most of the die-pressed samples developed a surface appearance - almost as if they had random "etch spots" or pits on the surface. This appearance completely disappeared after further removal of .005" - .010" material. Sample finishes on the order of ~ 10μ inch was obtained at the slower rate, and ~ 15μ inch at the faster rate.

b) <u>Electric Discharge Machining</u>

An equivalent quantity of the same size magnets were prepared at our facility by electric discharge machining (EDM). A Servomet Spark machine type SMD was used to reduce the diameters of the samples at two rates of material erosion. This machine produces a rapid series of spark discharges of controlled energy between the electrode tool and the work piece, in this case the test magnet. The sparks will erode the magnet by melting a small crater of metal and vaporizing the liquid (kerosene) in the vicinity of the tool. Vaporizing the liquid causes the area in the vicinity of the discharge to be purged of molten metal. The energy level employed and the frequency of discharges determines the material removal rate and the surface finish.

With proper tool design this servo-controlled machine can produce precision dimensioned pieces on hard-brittle materials such as rare earth-cobalt magnets. The only real practical drawback is the length of time to produce a piece if the work thickness is substantial.

Our experimental interest though is not to determine or prove whether this machine can work magnet surfaces, but to determine the resultant aging effects, if any, due to surface damage imparted to the magnets during the spark erosion process. If the melted surfaces impose macro or micro-strained layers or chemically affected layers to any significant depth, then the elevated temperature aging characteristic may also be affected.

From our previous experience in EDM machining of rare earth-cobalt permanent magnets we selected one spark rate setting of 4 for "lighter" working of each magnet and 3 for a "heavier" working. The working tool was actually tubular and "cored" the test magnet from the larger piece. Rate 4 produced a sample cored from a 0. 200" thick piece in about 4 1/4 hours, or ~0. 0008" per minute with about a 30μ inch finish. Rate 3 produced a sample in about 3 1/2 hours, or about 0. 001" per minute, with about a 50μ inch finish.

The only difficulty encountered in this operation other than the length of time involved, was that 2 (out of 10) thin GE samples (0. 500" diameter 0. 025" thick" cracked while EDM machining at each rate. These thin isopressed samples may be highly stressed and require only a small localized shock or stress to fracture them.

c) Surface Ground Raytheon Samples

To produce diepressed samples of the required length to diameter ratios to compare with isopressed samples in the elevated temperature evaluations, we proceeded to grind a sufficient number of Raytheon's samples (L/D = 0. 001"/0. 250" to the required thickness with the following results.

A total of 40 samples were selected based on the respective magnetic properties of each piece. Specific grinding procedures based on the optimum technique described by GE were determined by our machinists.[3] The equipment used was a standard Taft-Pierce horizontal surface grinder with a 1/2" wide aluminum oxide wheel (32A60-K5VBE) rotating at 3600 revolutions per minute. A down-feed of 0.0005", 0.050" crossfeed per pass, and surface speed ~20 feet per minute was used. Cimco 30 coolant solvent was used during the operation. Magnet pieces were blocked (restricted by ferromagnetic shims) and held on a magnetic parallel which was mounted on a magnetic chuck.

Four samples were ground as a preliminary exercise to preparing actual test samples. A close examination (Figure 19) of these samples after the removal of ~0.010" of material revealed that various die-pressing defects can be present that are not in evidence at the original surface. There were voids (in one case quite severe), minor edge cracks running radially inward, and semicircular edge cracks.

In grinding the actual group of test samples to size, material was removed in 4 steps at the rate of 0.0005" vertical feed per pass for the first 0.025" and the final 0.0125" at 0.0002" vertical feed, from each side. After each step the samples were examined carefully and surface conditions noted. The general observations at the end of each of these steps are as follows:

<u>Step 1.</u> After removal of 0.025" - one side"

4 samples - no noticeable cracks

1 sample - fine circular arc crack across entire surface

2 samples - minor trace surface crack or void

32 samples - small edge cracks

<u>Step 2.</u> After removal of last 0.0125" on the same side"

3 samples - no visible cracks

1 sample - large pit ground away

35 samples - small edge and circular cracks

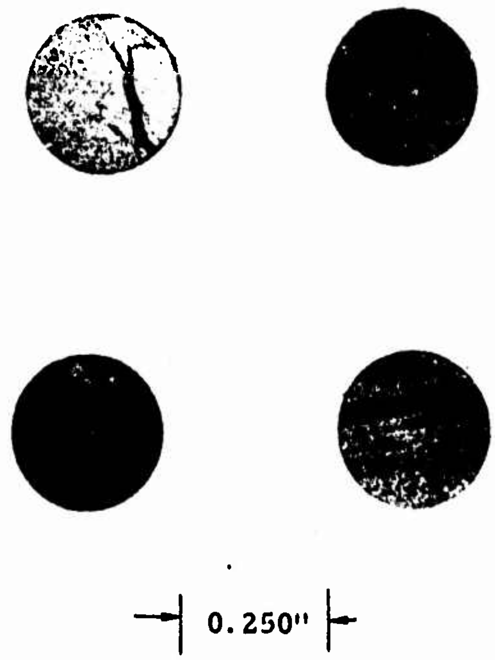

Figure 19. Internal Macro-Cracks Observed in Diepressed Test Magnets, After Removal of ~ 0.010 inches of Surface Material.

Step 3. Samples turned over and 0.025" removed on opposite side:

Surface condition observed this side:

4 samples - no visible cracks

3 samples - severe cracks or voids

6 samples - medium size cracks

25 samples - small cracks

2 samples - cracked through - one previously noted, another which had 2 radial edge cracks intercept at an angle

Step 4. Sample ground to 0.025" thickness - additional 0.0125" removed on same side:

Samples removed and inspected:

2 samples - no visible cracks either side

4 samples - cracked through

16 samples - edge cracks, both sides - same place

7 samples - circular edge crack both sides

7 samples - edge crack one side

2 samples - circular edge crack one side

2 samples - more than 1 edge crack each side

The grinding then, resulted in a 10% loss by cracks exposed in reducing the thickness of die-pressed samples. Inspection indicated a high probability of 42.5% (17 out of 40) of the samples would crack if further grinding was attempted to obtain $B/H = 1/8$ ($L/D = 0.0125''/0.250''$) specimens. Rather than risk the high loss of test samples due to further machining, it was decided to select the best of the samples for use as $B/H = 1/4$ test magnets in the aging evaluations from 150° to 250°C.

3. DISCUSSION OF THE AGING RESULTS ON MACHINED SAMPLES

a) Centerless Ground and Electric Discharge Machined Samples

In Tables 15 to 20, we present the results of data observed on GE and Raytheon magnets machined by both methods from larger stock pieces.

The initial properties listed in the table are the property values by which the samples were assigned in similar pairs prior to machining. After pulse charging (twice at 60 kOe) and remeasuring each sample with the proper hysteresigraph coil an examination of the general demagnetization curve show no sign of contour change indicating surface damage due to machining by either method. We do observe a small difference in remanent induction values for some but not all samples measured. For most samples regardless of the treatment or source, B_r changed less than ±1%, a few samples covered a range of -2.5 to + [illegible].6 percent. There are many factors which may have influenced the range of the measured values; a small air gap or poor sample contact with the pole faces of the electromagnet, small error in measuring the diameter and correcting for the sample area before or after machining, a small amount of drift in the hysteresigraph integrators, small edge chips not noted in the initial log when the samples were first measured. Any one or a combination of these factors could account for the changes observed.

The range of best and worst case effects of long-term accelerated air aging at temperatures from 150 to 250°C on the magnets machined by either process is shown in Figure 20, (B/H = 1/4), and Figure 21 (B/H = 2 1/2). A comparison of this data with preceding results of samples in the "as-received" condition can now be made.

1) <u>Permeance B/H = 1/4; 150°C Aging</u>

Referring to Tables 11 and 15 and the normalized curves shown in Figure 7b) and 20a), we observe some interesting results for the GE magnets. For the machined magnets the initial losses regardless of the surface removal rate, method, or original magnetic properties, now have a well defined range from 5.5 to 7.6 percent. The initial losses due to grinding were uniformly higher than EDM, but both methods yield lower losses than the sample tested in the as-received condition. The time to a 1 or 3 percent loss (referred to 15 minute values) is more uniformly shorter than those in the as-received condition. The total additional loss for the same time-temperature (~ 1935 hours at 150°C) history was also significantly higher.

TABLE 15

MACHINED SAMPLES, ACCELERATED AGING IN AIR AT 150°C,

B/H = 1/4

Sample Number	Initial Loss* (%)	Total Additional Loss: Loss ** (%)	Total Additional Loss: Time (Hours)	Time to Loss of 1% (Hours)	Time to Loss of 3% (Hours)	Initial Properties $_M H_c$ (kOe)	Initial Properties H_k (kOe)	Machining Method
A-622	7.3	4.23	1935	2	360	26.9	5.9	Ground @ Rate 1
A-621	7.6	3.64	1935	3.4	630	26.9	6.2	Ground @ Rate 2
A-629	7.4	3.82	1935	4.5	450	25.3	5.7	Ground @ Rate 1
A-626	7.6	3.99	1935	4	400	28.7	5.6	Ground @ Rate 2
A-641	6.3	3.40	1935	18	900	27.9	6.1	EDM @ Rate 4
A-638	6.[illegible]	3.07	1935	5	1600	27.4	6.5	EDM @ Rate 3
A-639	6.0	3.01	1935	15	1800	27.4	6.0	EDM @ Rate 4
A-618	5.5	2.98	1935	1.7	---	27.2	5.9	EDM @ Rate 3

*) Drop during first 15 minutes, expressed in % of initial value.
**) Drop from the value at 15 minutes, expressed in % of this value.

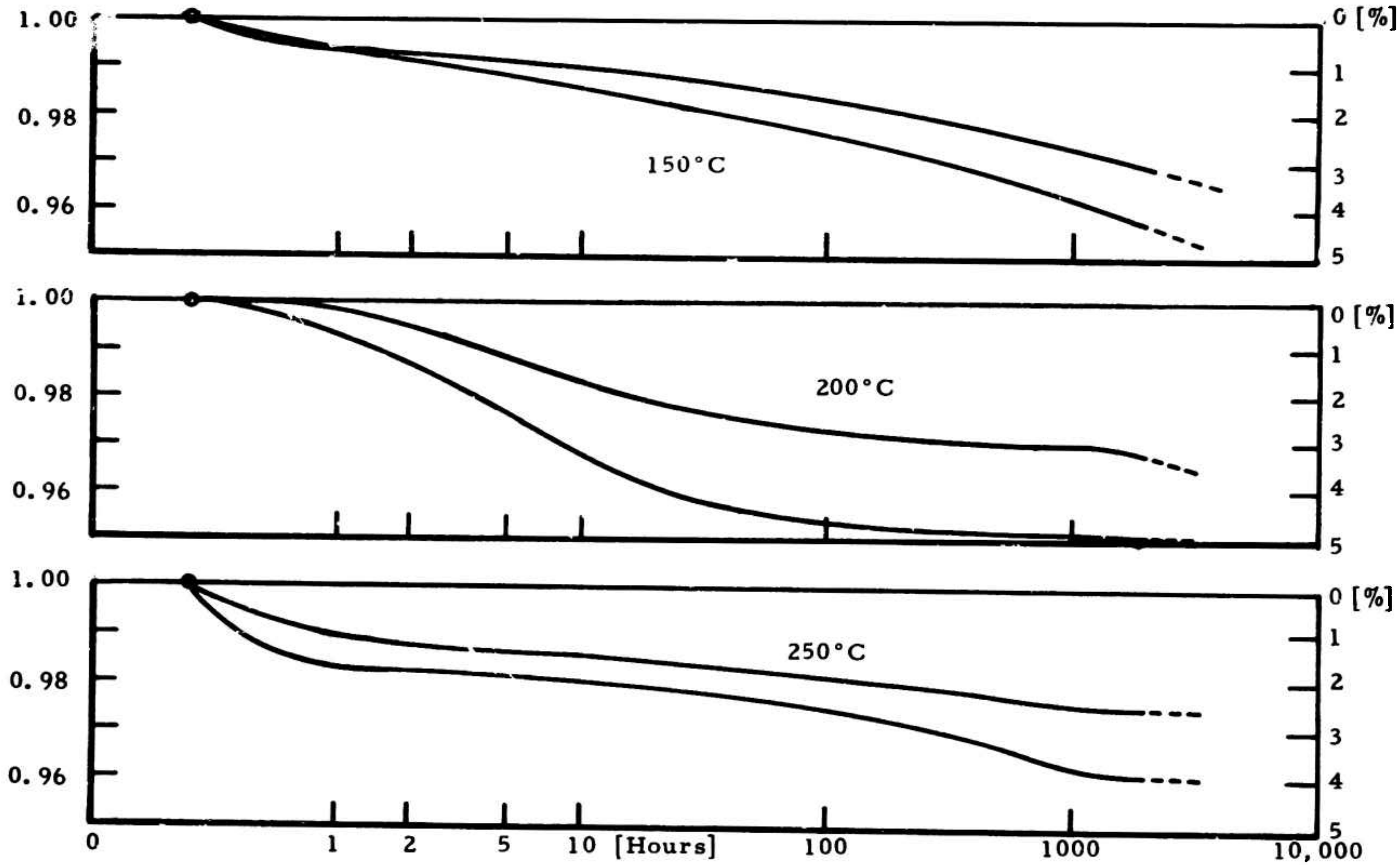

Figure 20. Machined Samples, Accelerated Air Aging, B/H = 1/4.
(a) 150°C (b) 200°C (c) 250°C

2) <u>Permeance of B/H = 1/4; 200°C Aging</u>

Referring to Tables 9 and 16, and the normalized curves of Figures 6a) and 20b), we can observe that the initial loss 5 to 8. 7%) is again more uniformly defined although of approximately the same magnitude. Again, the time to a 1 and 3 percent additional loss is still shorter for the samples we machined. Finally, the total additional loss over the evaluation interval (~ 1842 hours at 200°C) is still much greater than for those in the as-received condition.

3) <u>Permeance B/H = 1/4, 250°C Aging</u>

Referring to Tables 7 and 17, and the normalized curves of Figures 4a) and 20c), we still observe the same trend of a more uniform range of initial loss (~ 105 to 14. 4%) and about the same magnitude. In this case the time to a 1% loss was shorter as before but the loss to 3% was slightly longer. However, the total loss at 1879 hours, is once again slightly higher than the samples in the as-received condition.

4) <u>Permeance B/H = 2 1/2; 150°C Aging</u>

Referring to Tables 10 and 18, and the normalized curves shown in Figures 7a) and 21a), we can make a general comparison of the effects of machining on aging characteristics even though there is a large disparity between the B/H numbers. For these longer samples of the same diameter (of each manufacture) we find that the initial losses were 2. 5 percent or less (with few exceptions). Approximately 69% were 1. 8% or lower, which is comparable to that indicated by the samples in Table 10. The time to a 1 or 3% loss has about the same nominal range of hours. It does appear though the diepressed samples have a shorter time to a loss of 1% than the isopressed samples. There does not appear to be any difference (at this temperature) attributable to the method of machining even though the surface areas worked were much larger. The same appraisal applies to the total additional losses incurred over a comparable time span.

TABLE 16

MACHINED SAMPLES, ACCELERATED AGING IN AIR AT 200°C,

B/H = 1/4

Sample Number	Initial Loss* (%)	Total Additional Loss: Loss ** (%)	Total Additional Loss: Time (Hours)	Time to Loss of 1% (Hours)	Time to Loss of 3% (Hours)	Initial Properties: $_MH_c$ (kOe)	Initial Properties: H_k (kOe)	Machining Method
A-609	8.7	3.83	1842	2.2	15	26.6	7.4	Ground @Rate 1
A-640	8.4	3.32	1842	2.3	90	24.9	6.7	Ground @ Rate 2
A-606	7.7	3.78	1842	2	38	25.8	7.4	Ground @ Rate 1
A-601	8.1	3.83	1842	2	3.8	26.4	7.3	Ground @ Rate 2
A-635	5.0	3.13	1842	3.4	1000	26.8	6.6	EDM @ Rate 4
A-636	5.6	3.46	1842	2.3	140	25.6	7.8	EDM @ Rate 3
A-637	6.7	4.92	1842	1.5	8.5	25.7	5.5	EDM @ Rate 4
A-615	7.8	4.05	1842	2	21	26.1	6.1	EDM @ Rate 3

*) Drop during first 15 minutes, expressed in % of initial value.
**) Drop from the value at 15 minutes, expressed in % of this value.

TABLE 17

MACHINED SAMPLES, ACCELERATED AGING IN AIR AT 250°C,

B/H = 1/4

Sample Number	Initial Loss* (%)	Total Additional Loss: Loss ** (%)	Total Additional Loss: Time (Hours)	Time to Loss of: 1% (Hours)	Time to Loss of: 3% (Hours)	Initial Properties: $_MH_c$ (kOe)	Initial Properties: H_k (kOe)	Machining Method
A-617	13.9	1.81	1879	8	---	27.8	5.5	Ground @ Rate 1
A-607	13.5	3.15	1879	1.5	1875	27.1	6.6	Ground @ Rate 2
A-610	14.37	4.04	1879	0.78	790	25.5	6.0	Ground @ Rate 1
A-630	13.89	3.6	1879	1.1	---	27.2	7.0	Ground @ Rate 2
A-634	12.7	3.54	320***	0.55	---	27.4	7.0	EDM @ Rate 4
A-632	12.56	2.43	1879	0.75	---	27.6	6.6	EDM @ Rate 3
A-619	13.75	2.86	1879	0.73	---	26.6	6.6	EDM @ Rate 4
A-612	10.50	1.79	1879	1.20	---	28.1	7.9	EDM @ Rate 3

*) Drop during first 15 minutes, expressed in % of initial value.

**) Drop from the value at 15 minutes, expressed in % of this value.

***) Sample Cracked after 320 hours during test.

TABLE 18

MACHINED SAMPLES, ACCELERATED AGING IN AIR AT 150 °C

B/H = 2.5

Sample Number	Initial Loss* (%)	Total Additional Loss: Loss** (%)	Total Additional Loss: Time (Hours)	Time to Loss of: 1% (Hours)	Time to Loss of: 3% (Hours)	Initial Properties***: $_{M}H_{c}$ (kOe)	Initial Properties***: H_k (kOe)	Machining Method
A-502	1.5	1.44	1935	210	--	51.7	12.7	Ground @ Rate 1
A-537	1.7	1.31	1935	450	--	48.8	10.9	Ground @ Rate 2
A-514	1.2	1.45	1935	210	--	43.3	11.8	Ground @ Rate 1
A-515	1.8	0.66	1935	--	--	43.8	9.6	Ground @ Rate 2
A-508	1.1	1.39	1935	270	--	61.0	13.3	EDM @ Rate 4
A-512	1.8	0.94	1935	--	--	58.4	15.0	EDM @ Rate 3
A-530	1.6	1.37	1935	300	--	49.6	7.9	EDM @ Rate 4
A-529	2.2	1.59	1935	110	--	45.4	8.7	EDM @ Rate 3
B-526	1.0	1.82	1935	37	--	52.4	10.2	Ground @ Rate 1
B-532	1.3	1.72	1935	60	--	44.7	11.3	Ground @ Rate 2
B-501	2.5	2.96	1935	16	--	33.6	5.2	Ground @ Rate 1
B-521	3.2	1.75	1935	36	--	33.5	4.4	Ground @ Rate 2
B-509	1.6	1.67	1935	110	--	33.3	7.0	EDM @ Rate 4
B-517	13.1	2.08	1935	29	--	33.1	6.1	EDM @ Rate 3
B-513	1.7	1.87	1935	62	--	31.8	5.5	EDM @ Rate 4
B-536	2.5	1.87	1935	50	--	31.8	5.2	EDM @ Rate 3

* Drop during first 15 minutes, expressed in % of initial value.
** Drop from the value at 15 minutes, expressed in % of this value.
*** Properties Before machining

5) Permeance B/H = 2 1/2, 200°C Aging

Referring to Tables 8 and 19, and the normalized curves shown in Figure 5 and 21b), we observe much the same general trends as at the lower temperature. Although the initial losses have approximately the same range, the additional time-loss characteristics to 1 and 3 are generally longer. The total loss for a comparable length of time (1842 hours) is much improved over the "as-received" samples. This may be attributed to the significantly larger volume of the longer machined magnets.

6) Permeance B/H = 2 1/2; 250°C Aging

Referring to Tables 6 and 20, and the curves shown in Figure 3 and 21c), we now observe that although the initial losses are ~1% higher than the previous level of temperature exposure (200°C), the range of losses is much narrower and at the lower end of the loss range observed for the "as-received" samples. The machined diepressed samples still show a greater initial loss than isopressed samples. The data also generally indicate that for the isopressed samples the time to a loss of 1 and 3% is longer with the ground samples. For the die pressed samples only the 3% loss-time is improved. In Table 20, we can also observe that there is a time-loss effect attributable to the method of machining with samples of both manufactures. It now appears that almost all of the samples machined by the EDM method are severely affected in short term stability time to 1% loss. The percent level of total loss is also slightly greater for samples EDM machined and exposed at this temperature. However, the total loss in OCRI is improved over the (B/H ≈ 1) samples in the "as-received" condition.

7) General Observations

Based on the data that we have tabulated and compared we can make the following general comments regarding the observable aging effects by these two methods of machining:

Both machining methods have narrowed the range of initial losses incurred regardless of the temperature assignment.

The initial losses on samples, L/D = 0.025"/0.250" (B/H ≈ 1/4), were greater for the centerless ground samples than those EDM machined.

TABLE 19

MACHINED SAMPLES, ACCELERATE AGING AT 200°C,

B/H = 2.5

Sample Number	Initial Loss* (%)	Total Additional Loss Loss** (%)	Total Additional Loss Time (Hours)	Time to Loss of 1% (Hours)	Time to Loss of 3% (Hours)	Initial Properties $_{M}H_{c}$ (kOe)	Initial Properties H_k (kOe)	Machining Method
A-536	2.6	1.11	1842	100	--	44.4	11.8	Ground @ Rate 1
A-539	2.7	1.37	1842	62	--	46.4	11.3	Ground @ Rate 2
A-501	3.0	1.01	1842	1840	--	49.1	10.4	Ground @ Rate 1
A-517	2.7	1.11	1842	900	--	50.9	13.1	Ground @ Rate 2
A-532	1.9	1.26	1842	150	--	49.5	8.5	EDM @ Rate 4
A-533	2.3	1.49	1842	40	--	49.6	11.6	EDM @ Rate 3
A-518	2.5	0.97	1842	--	--	49.1	7.8	EDM @ Rate 4
A-522	3.2	1.12	1842	50	--	44.2	9.5	EDM @ Rate 3
B-500	2.8	1.44	1842	135	--	35.3	8.2	Ground @ Rate 1
B-525	2.8	1.36	1842	22	--	35.2	5.7	Ground @ Rate 2
B-512	4.0	2.15	1842	4.5	--	32.8	5.3	Ground @ Rate 1
B-528	5.6	1.37	1842	26	--	33.7	4.4	Ground @ Rate 2
B-503	4.8	1.44	1842	22	--	35.5	4.9	EDM @ Rate 4
B-540	4.1	2.31	1842	4.5	--	34.6	5.2	EDM @ Rate 3
B-516	4.1	2.00	1842	7	--	31.8	5.0	EDM @ Rate 4
B-508	3.1	2.29	1842	7.5	--	32.6	5.8	EDM @ Rate 3

* Drop during first 15 minutes, expressed in % of initial value.

** Drop from the value at 15 minutes, expressed in % of this value.

TABLE 20

MACHINED SAMPLES, ACCELERATED AGING IN AIR AT 250°C

B/H = 2.5

Sample Number	Initial Loss * (%)	Total Additional Loss: Loss ** (%)	Total Additional Loss: Time (Hours)	Time to Loss of 1% (Hours)	Time to Loss of 3% (Hours)	Initial Properties: $_MH_c$ (kOe)	Initial Properties: H_k (kOe)	Machining Method
A-531	3.6	1.44	1879	700	---	55.7	13.4	Ground @ Rate 1
A-534	3.5	1.71	1879	700	---	49.8	14.0	Ground @ Rate 2
A-509	3.9	1.89	1879	490	---	53.0	12.3	Ground @ Rate 1
A-523	4.0	2.39	1879	200	---	46.4	12.5	Ground @ Rate 2
A-521	4.5	2.15	1879	4	---	51.2	10.9	EDM @ Rate 4
A-535	3.4	2.18	1879	30	---	49.6	10.5	EDM @ Rate 3
A-513	3.2	2.00	1879	60	---	53.0	10.6	EDM @ Rate 4
A-510	4.2	1.00	1879	1879	---	53.0	10.0	EDM @ Rate 3
B-543	6.4	1.28	1879	25	---	35.8	5.7	Ground @ Rate 1
B-534	6.5	1.22	1879	30	---	35.8	5.5	Ground @ Rate 2
B-511	8.2	1.04	1879	1800	---	32.4	4.4	Ground @ Rate 1
B-502	6.8	1.36	1879	25	---	32.5	5.3	Ground @ Rate 2
B-518	3.1	1.53	1879	5	---	33.2	7.8	EDM @ Rate 4
B-515	3.3	1.71	1879	3	---	33.2	8.1	EDM @ Rate 3
B-522	4.3	1.75	1879	2	---	31.7	6.1	EDM @ Rate 4
B-514	5.5	1.62	1879	3	---	32.0	5.5	EDM @ Rate 3

*) Drop during first 15 minutes, expressed in % of initial value.

**) Drop from the value at 15 minutes, expressed in % of this value.

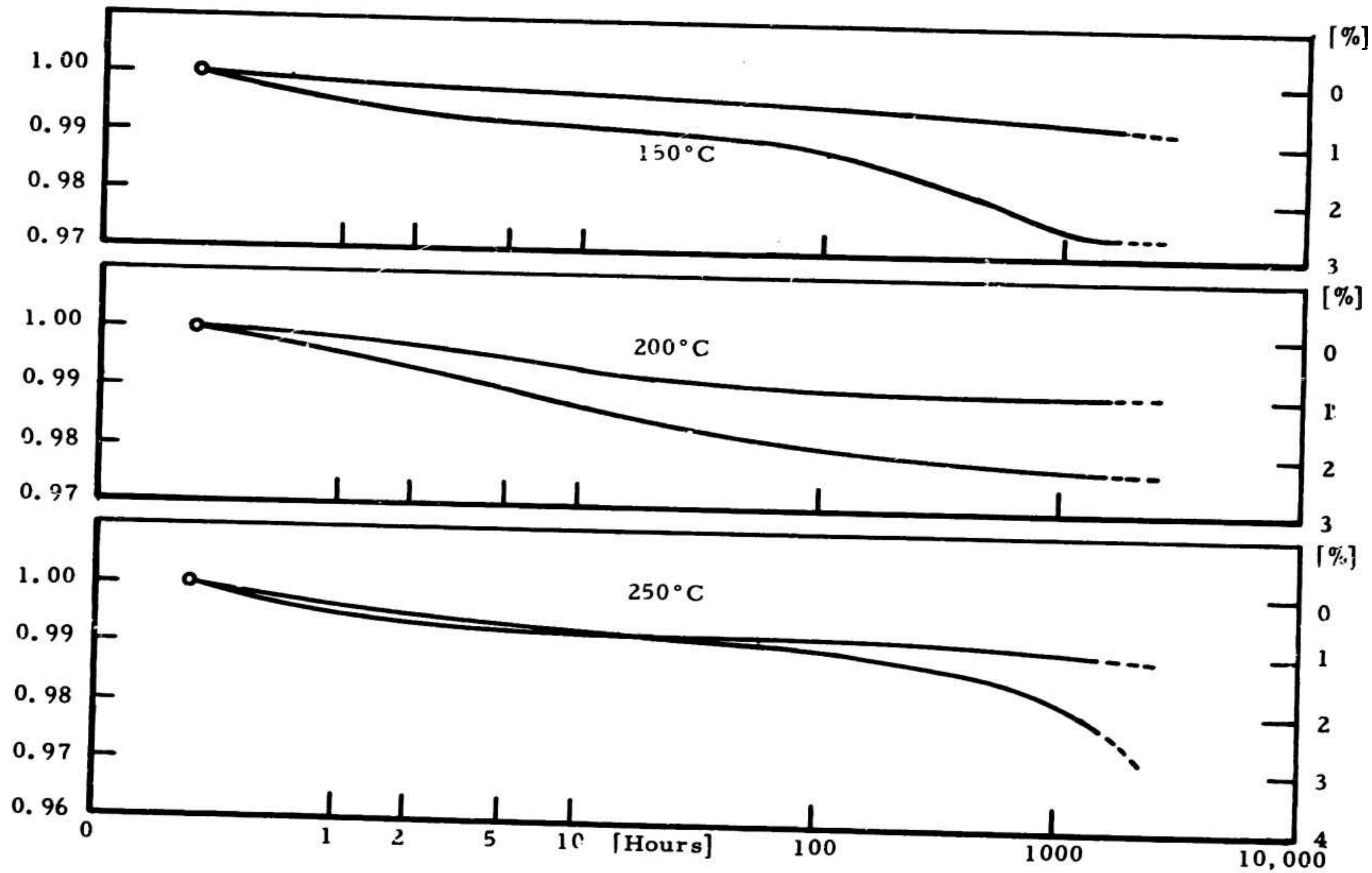

Figure 21. Machined Samples, Accelerated Air Aging, B/H = 2-1/2
(a) 150°C (b) 200°C (c) 250°C

The initial loss range for B/H = 1/4 samples was less for the lower temperature exposure of 150°C, and approximately the same for the 200°C and 250°C exposure when compared to the "as-received" sample losses.

The additional time to losses of 1% was also shorter at 250°C but longer to 3%.

The total losses observed for comparable lengths of time were higher for the machined samples than the "as-received" samples of B/H = 1/4.

The more massive machined samples (B/H >1) exhibit the same trend of a much narrower spread in initial losses.

The initial losses of machined samples are of the same order at 150° to 200°C, and only 1% higher at 250°C than "as-received" B/H ≈ 1 samples.

The time for 1 and 3% losses at the lower temperature exposure (150°C) are about the same for B/H = 2 1/2; in comparison to B/H ≈1 samples.

The time for losses of 1 and 3% at 200°C are longer.

At 250°C the method of machining used becomes apparent in the time rate of loss. In this experiment the grinding method yielded the best results. Isopressed sample stability was longer for a 1 and 3% loss, and only longer for the 3% level with the diepressed samples.

The total losses for the B/H = 2 1/2 samples is about the same at 150°C, but is substantially improved at 200°C and 250°C.

From these general comments and observations it would appear that the machining methods used have produced one common affect on samples of either size or manufacture, that is an improvement in the degree of range of the initial loss percentages. Whether this is due to the removal of some oxide surface layer, or slight preaging through material removal is not known.

It also appears that the results obtained at specific temperatures are directly dependent on the surface to volume ratio of the samples examined. On the one hand we had samples for which only a minor amount of surface was machined and another in which the major portion of the surface was worked by either method.

b) Surface Ground Samples

In Table 21 and Figures 22 and 23 we present the results of Raytheon samples surface ground to a size for B/H = 1/4 permeance. In every case these samples had some form of visible macro-crack or voids on one side or both after the grinding operation. For comparable exposure times at each respective temperature these samples appear to degrade much more severely than isopressed samples of similar geometry as shown in Tables 7, 9 and 11. These samples were rather well matched up regarding their magnetic properties, and comparable to those of GE manufacture. In some cases the initial losses were quite similar for each pair but generally higher than the isopressed samples. In most cases the time-loss to 1 and 3%, and the total loss were also similar in the group. These observations can best be attributed to the poor surface integrity of the magnet sample after the removal of a significant amount of material from each surface. One would certainly expect a rapid oxidation of the surfaces exposed at these temperatures.

TABLE 21

MACHINED SAMPLES ACCELERATED AIR AGING, B/H = 1/4

Sample Number	Initial Loss* (%)	Total Additional Loss: Loss** (%)	Total Additional Loss: Time (Hours)	Time to Loss of 1% (Hrs.)	Time to Loss of 3% (Hrs.)	Time to Loss of 5% (Hrs.)	Initial Properties $_MH_c$ (kOe)	Initial Properties H_k (kOe)	Machining Method	Evaluation Temp. (°C)
B-212	8.5	4.96	1935	90	90	---	27.0	3.8	Surface Ground	150
B-223	10.0	4.70	1935	0.8	270	---	28.2	3.5	"	150
B-200	17.0	7.42	1935	0.8	12.5	142	23.4	2.6	"	150
B-201	14.6	5.79	1202***	1.5	33	520	23.5	2.8	"	150
B-203	10.7	4.62	1842	0.9	17	---	26.5	4.4	"	200
B-217	8.7	4.15	1842	16.5	32	---	26.1	4.3	"	200
B-237	22.0	7.18	1842	0.8	4	19	22.7	2.6	"	200
B-202	21.9	7.83	1842	0.5	2.5	14	22.7	2.7	"	200
B-208	16.0	5.70	1879	0.5	8	580	24.7	3.2	"	250
B-214	16.2	6.06	1879	1.2	5	300	24.6	3.3	"	250
B-207	16.1	5.5	1879	0.5	9	950	24.3	3.8	"	250
B-213	16.4	5.0	1879	0.6	72	1800	24.2	3.8	"	250

* Drop during first 15 minutes, expressed in % of initial value.

** Drop from the value at 15 minutes, expressed in % of this value.

*** Sample cracked after measurement

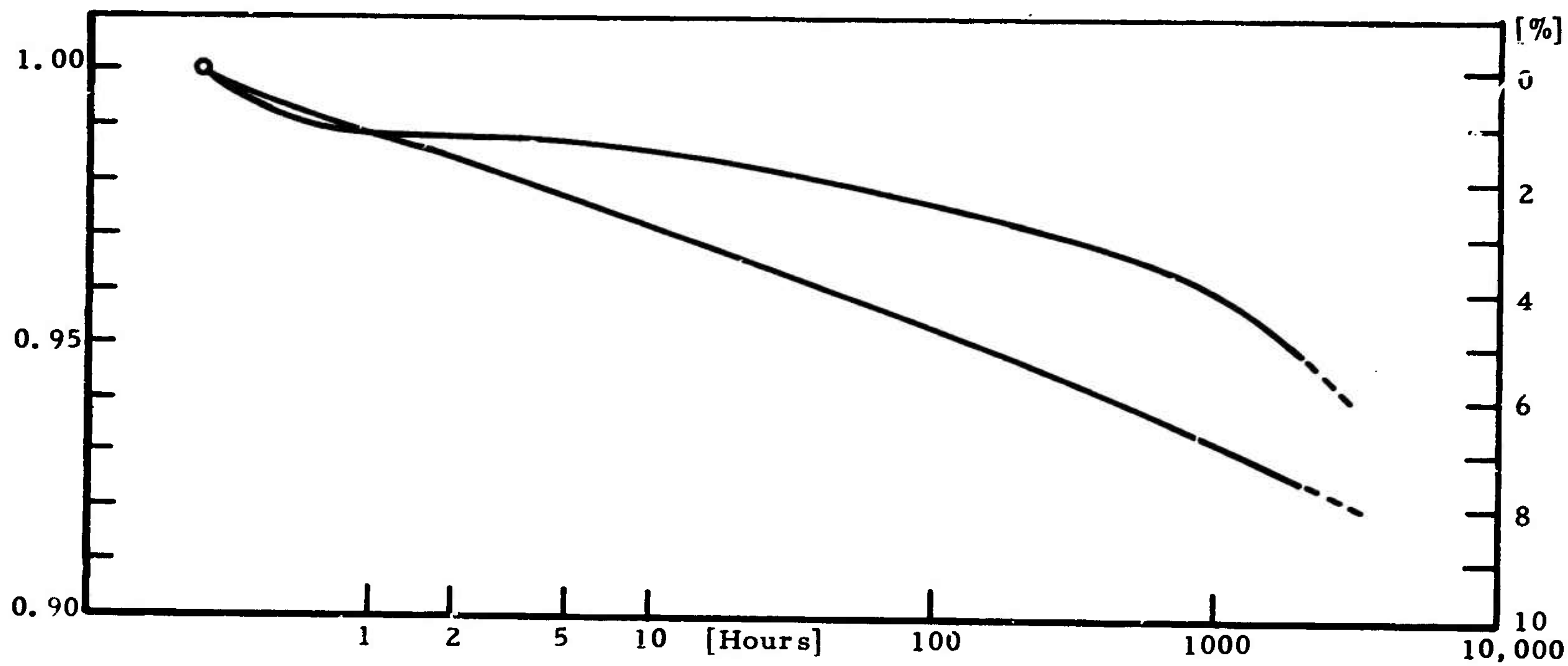

Figure 22. Machined Samples, Raytheon B/H = 1/4, Accelerated Air Aging at 150°C.

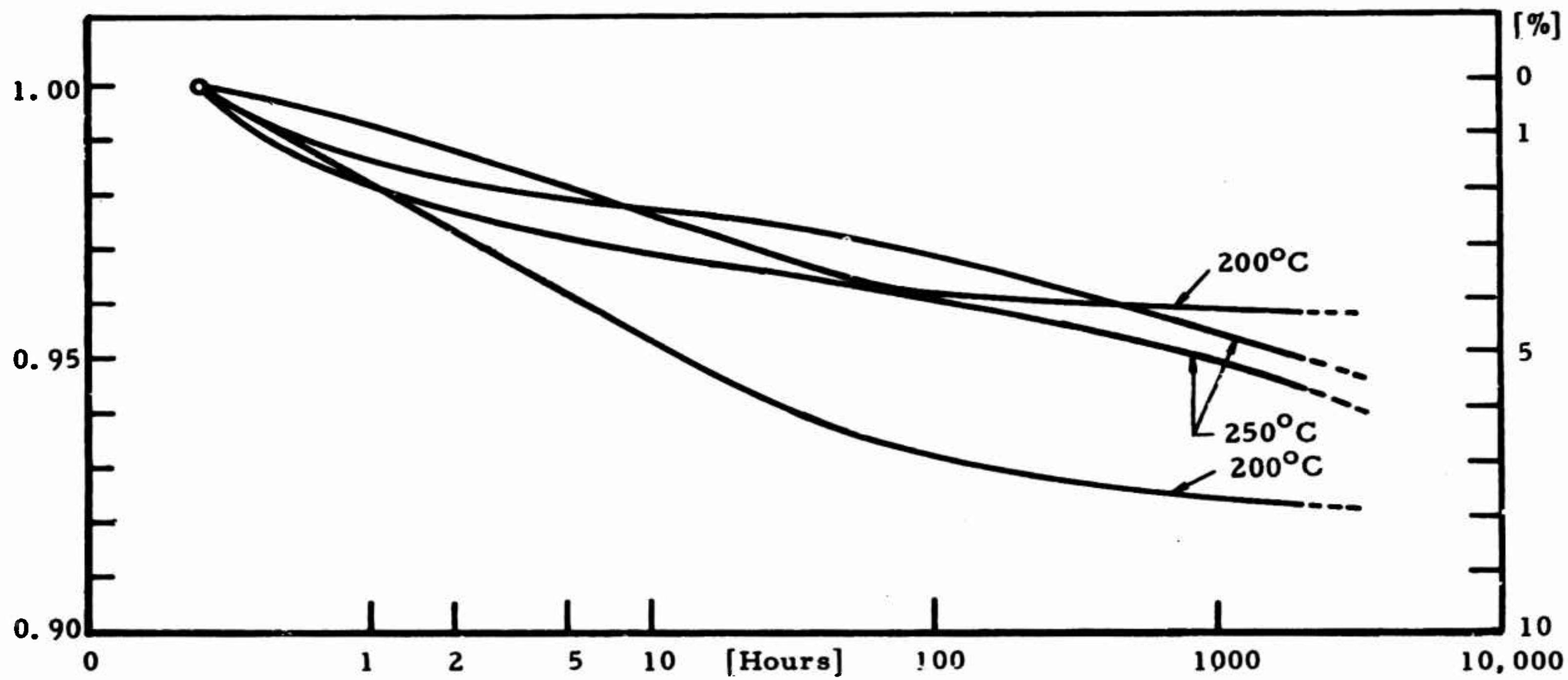

Figure 23. Machined Samples, Raytheon B/H = 1/4, Accelerated Air Aging at 200° and 250°C.

SECTION V

COMPRESSIVE STRENGTH - THERMAL AGING EFFECTS

1. PURPOSE OF THIS EXPERIMENT

In certain applications of permanent magnets there may be encountered a combination of compressive stresses, shock, tension, etc., in addition to thermal environments which may seriously alter the performance of a magnet. To simply examine the combination of two of these environments, - compressive loading and thermal exposure, we investigated first, the physical compressive strengths at temperature from 22°C to 250°C, and then long-term compressive stress vs. temperatures from 150°C to 250°C.

2. EXPERIMENTAL PROCEDURE

a) Compressive Stress Tests, 22°C to 250°C

To determine the relative compressive strength of the die pressed and isopressed samples as a function of temperature, we assigned samples (L/D = 0.100"/0.250") from our supply of stock magnets. The basis of selecting these samples was primarily the magnitude of the $_MH_c$ value we recorded. The assumption being that the higher $_MH_c$ is the higher the density, hence better structural integrity. Although our questionable density measurements do not support this we felt it would be interesting to see if some trend could be observed.

The compressive tests were conducted using an Instron Universal Tester, with automatic load pacer, load cell-recording system, and temperature chamber. This system also has ten-step zero suppression, to greatly magnify selected portions of the stress-strain curve for a detailed analysis. Load reading accuracy is certified ±1% on any range. The samples selected were examined with an optical comparator to insure that the surfaces were essentially plane-parallel and smooth. Each sample listed in Table 22 was then placed in the testing apparatus protected by .005" tantalum spacer shims (to avoid minute surface irregularities of either a sample or the ram head, surfaces, and stabilized at the test temperature. At temperature, the load pacer was operated at 33 microinches per second, and the sample compressively stressed while signs of internal stress and ultimate sample failure were observed and recorded. In most cases small dips in the stress curves attributed to internal faults were observed at low loads before ultimate sample failure occurred.

The data listed in Table 22 under the column heading "Ultimate Stress" has been corrected for sample cross-sectional area. In each case the value listed under "Load" is the point where maximum stress change occurred in the sample. In some tests a small chip may shear off the sample first, for others the failure was instantaneous and resulted in a complete disintegration of the sample structure.

We observe a wide range of ultimate stress values for the isopressed samples, ranging from approximately 14 to 165 kpsi. Most of these samples failed above ~35 kpsi. The diepressed samples range from 39 to 104 kpsi, with most of them failing above ~50 kpsi. Brittle materials such as these magnets are, usually exhibit a high degree of scatter in compressive failure strength, as we have seen.

There does not appear to be any indication of a preferential compressive stress-temperature level, at least under static conditions.

b) <u>Long Term Compressive Stress - Thermal Tests</u>

For this experiment we prescreened a sufficient number of samples of each supplier by compressive loading to 10 kpsi. If no indication of internal faults were observed then the sample was allocated to a particular elevated temperature test.

Three fixtures were designed and constructed to provide a compressive loading on the samples when mounted in standard Creep-Rupture Test Stands. Each stand has a vertical tubular furnace mounted for easy

TABLE 22

COMPRESSIVE STRESS VERSUS TEMPERATURE DATA

SAMPLE L/D = 0.100"/0.250"

Sample Number	Evaluation Temperature (°C)	Coercive Force-$_MH_c$ (kOe)	Density (g/cm^3)	Load (lbs)	Ultimate Stress (10^3 psi)
A-24	22	47.0	8.6	2725	55.5
A-28	22	53.0	8.5	1800	35.5
A-55	22	34.7	8.5	7700	151.9
B-22	22	37.3	8.3	4450	89.2
B-23	22	32.9	7.9	2825	57.1
B-50	22	36.5	9.8	3575	77.2
A-34	150	51.1	8.8	7950	165.9
A-54	150	30.4	8.6	680	14.4
A-65	150	50.2	9.4	5250	110.4
B-30	150	30.9	8.8	5000	101.8
B-44	150	37.3	7.8	2550	50.7
B-55	150	34.2	8.2	1900	39.0
A-26	200	53.6	8.9	3450	70.3
A-36	200	55.2	8.7	3540	72.7
A-51	200	44.2	8.1	2100	43.8
B-49	200	37.1	8.3	5150	104.0
B-56	200	32.6	8.3	4150	85.2
B-57	200	35.7	9.0	3100	63.1
A-50	250	46.9	8.1	3500	73.6
A-56	250	40.9	8.3	4390	88.0
A-60	250	36.3	8.3	4550	95.7
B-19	250	36.9	8.6	4390	85.9
B-59	250	36.1	8.8	3750	77.6
B-65	250	33.7	8.8	4430	91.7

access to the specimen area. Each furnace was set for one of the desired temperatures 150°C, 200°C or 250°C. The test samples (OCRI) were measured before and after prescreening, and then placed in the loading fixture with protective tantalum shims. Actual sample loading was through a stainless steel piston slightly larger in diameter than the samples. After the samples are positioned, dead weight loading was applied for an equivalent stress of 10 kpsi on the sample. The furnaces would then be put in place and monitored until the system stabilized. At intervals, the samples were off-loaded, removed and OCRI measured at room temperature. Sufficient data was obtained to compare the results with stress-free aging characteristics. As noted in Table 23, a few of the isopressed samples fractured after a few hundred hours of aging either during the test interval, in removal from the fixture with aluminum tweezers (shaped to fit the sample diameter), or during the placement of the sample in the measurement fixture with the same tweezers. It appeared that the slightest change in pressure or rapid cooling was sufficient to cause the sample to fracture almost spontaneously.

3. RESULTS OF COMPRESSIVE STRESS-THERMAL AGING TESTS

The data measured in this experiment is presented in Table 23 and illustrated in Figure 24. If these data are referred to that of Table 10 and Figure 7a), (air aging at 150°C) we observe that the best case for samples under either condition is about the same, but the worst case (compression loaded sample) is much more drastic. This sample actually sustained very little loss in the first two hours and the next 8 hours the OCRI drops ~2 1/2 percent to a plateau for approximately 200 hours before undergoing another sustained drastic change. All the samples but one behaved rather comparable to the unstressed samples at this temperature.

At 200°C, if we refer to the data in Table 6 and Figure 5, and compare the results presented in Table 23 and Figure 24, we can observe that the best long-term aging was experienced with a compressively stressed sample. This best case result was 100% more stable than for an unloaded counterpart.

TABLE 23. COMPRESSIVE LOADING - THERMAL AGED SAMPLES, B/H = 1

Sample Number	Initial Loss* (%)	Total Additional Loss Loss** (%)	Time (Hours)	Time to Loss Of 1% (Hrs.)	3% (Hrs.)	5% (Hrs.)	Initial Properties $_MH_c$ (kOe)	H_k (kOe)	Evaluation Temperature (°C)
A-89	6.4	0.71	2932	---	---	---	49.4	10.7	150
A-74	1.00	1.10	336***	300	---	---	36.7	9.3	150
A-83	0.80	2.40	2594	560	---	---	46.1	9.2	150
B-33	0.12	6.07	3034	8.8	105	910	33.2	6.1	150
B-63	0.18	2.54	3034	3.5	---	---	26.5	10.3	150
A-58	0.76	3.09	3211	130	2400	---	42.5	10.0	200
A-49	0.80	3.30	300***	4	108	---	37.3	8.3	200
A-72	1.08	1.58	2765	340	---	---	36.8	10.6	200
B-24	1.74	3.97	3211	6.3	560	---	36.5	5.7	200
B-60	6.0	7.39	3211	3	7.5	43	29.5	5.1	200
A-59	4.6	1.5	194***	60	---	---	48.7	8.9	250
A-88	2.4	5.01	2318	370	1400	2310	49.4	11.2	250
A-75	4.9	1.00	194***	194	---	---	35.6	6.1	250
A-80	1.8	3.81	2318	310	1600	---	37.3	6.1	250
B-84	4.8	4.05	2512	134	1470	---	41.1	7.9	250
B-61	6.5	4.23	2512	94	1120	---	32.9	6.8	250

*) Drop during first 2 hours, expressed in % of initial value.
**) Drop from the value at 2 hours, expressed in % of this value.
***) Sample cracked during test exposure, or measurement.

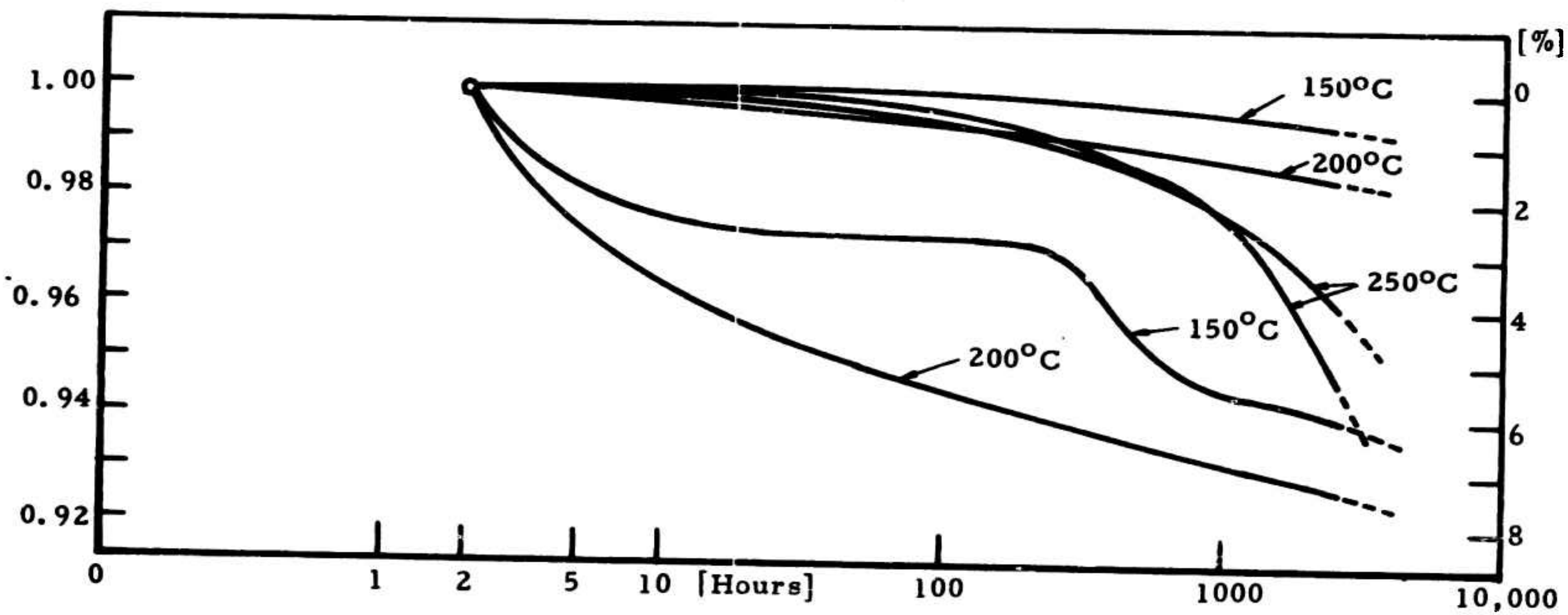

Figure 24. Compressive Stress - Elevated Temperature Air Aging, 150°C, 200°C and 250°C. B/H = 1.

Again, all the other samples but one are comparable to the unloaded samples. The worst case shows an extremely poor short term stability right at the beginning of the test.

At 250°C, the best case for the unstressed samples (Table 6, Figure 3) is $\sim$100 percent more stable than the stressed sample for the sample exposure time. The worst case loss for the stressed sample however is only about 66 percent of the unstressed sample worst case for a comparable time interval.

One peculiarity observed with the stressed samples is that the initial losses sustained by the samples in a 2 hour interval is significantly lower (with one exception in each case) than the "as received" samples in the first 15 minute interval, and unstressed. Whether this is attributable to the prestressing of the samples, which is highly unlikely, or strictly due to the chance of selecting samples which might behave more favorably in a given test, is uncertain.

One possible explanation is that the magnet samples in the unstressed air aged studies were rapidly brought to temperature in just a few short minutes during the first few hours of accumulated time. As we have previously reported[1] the sample aging was started in Driblock heaters. These units provided for quick removal and insertion of the magnets with a minimum of thermal recovery time to set temperature.

In the compressive loading tests, the repositioning and recovery of the furnace set temperature takes on the order of 30 to 60 minutes once it is in place. The rate of initial thermal shock on the magnet may be a factor in the magnitude of the initial loss.

Generally speaking then it appears that with only one exception at each of the lower temperatures exposures the samples age in a similar manner to the unstressed samples. At the higher temperature the best stressed samples appear to age more rapidly than an unstressed sample. At the higher temperature a narrower range between characteristic aging curves for the stressed samples was observed with the samples tested.

SECTION VI

EFFECTS OF MAGNETIC FIELD CYCLING ON THE OPEN-CIRCUIT REMANENT FLUX

1. PURPOSE OF EXPERIMENT

In many applications of permanent magnets, such as motors and generators, certain conditions may occur which could produce severe demagnetizing fields. Many application designs using low coercivity magnets are therefore compromised or restricted in rating as a result, to minimize the effect of such fields.

In this experiment then we wished to observe the long-term stability of $SmCo_5$ magnets under the influence of alternating magnetic fields comparable to the magnitude of $_BH_c$ for each type of magnet.

2. EXPERIMENTAL PROCEDURE AND RESULTS

The test magnets were selected from the large second lot of stock samples from each supplier with a $B/H \approx 1$. In Figure 25 we illustrate the best match possible of B vs. H curves of 4 samples selected from each manufacturer.The values of $_BH_c$ and B_r are also listed in Table 24. With this information, an average open-circuit self-demagnetizing field value, $-H_d$, was determined for each group. The magnets were pulse charged again and the initial OCRI measured. Each sample group was then placed in a separate glass-epoxy holder and inserted in a fixed gap of an electromagnet with 3 inch polecap diameter.

All samples were in direct contact with the pole faces. It was then arranged to sweep the field of each of the two electromagnets sinusoidally at 1000 cycles per hour, with an amplitude of $\pm$ 0.86 H_d, symmetrically about a steady bias field of $-H_d$, the average open-circuit operating-point field for the respective sample group.

Although this magnetic cycling experiment was inspired by the idea to simulate the approximate (magnetic) operating conditions which a magnet experiences in a rotating electrical machine, it was necessary to establish

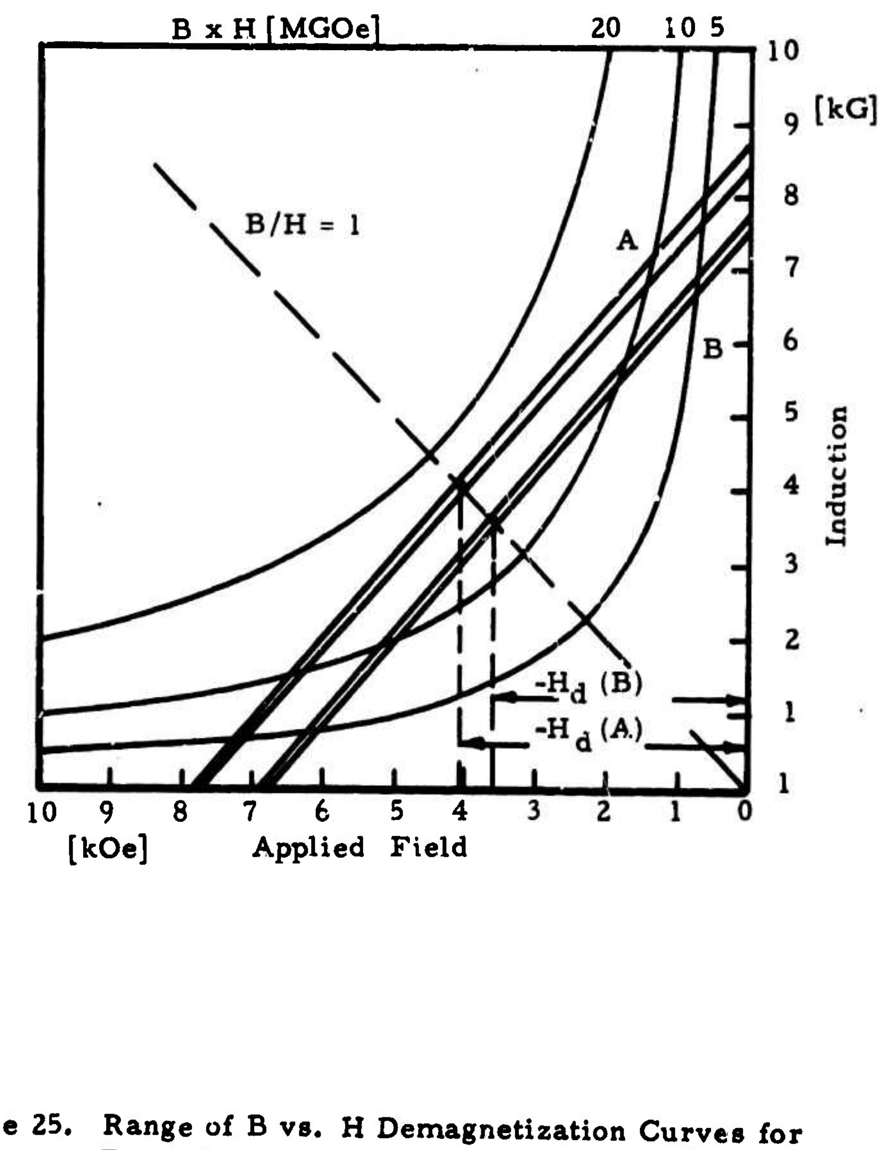

Figure 25. Range of B vs. H Demagnetization Curves for Field Cycling Effect Test.

simple and clearly defined cycling procedures to eliminate possible uncertainties about the meaning of the results. First, the frequency - - 1000 cycles per hour (cph), or 0.28 Hz - - was chosen low enough to eliminate problems with eddy-current shielding in the samples or pole pieces, and thus to ensure that all volume elements of all samples were subject to substantially the same instantaneous magnetic fields. Second, before the 1000 cph "a.c." exposure began, the samples were taken slowly and with manual field adjustment through several cycles and the open-circuit remanent induction was measured after each individual cycle. This allowed a separation between the "trivial knockdown" of the OCRI during the initial half cycle which is due to the nonlinearity of the second-quadrant demagnetization curve, any short-term flux reduction that may occur during the first few full cycles after that, and genuine long-term "aging effects" above and beyond the previously investigated static natural air aging which may be caused by the often-repeated magnetic cycling.

The cycling and measuring procedures used were as follows. The sequence of magnetization states is depicted in the Figures; the differences between the various states and recoil loops are highly exaggerated for the sake of clarity.

A. Slow cycling between, approximately, the open-circuit operating point and the short-circuited condition. (See Figure 26a)

1) OCRI value corresponding to $B_{d,1}$ after pulse charging of the sample was measured (operating point O).
2) Sample was put into gap of electro-magnet yoke (point 1') the field was swept $H_a = 0 \rightarrow -H_{d,av} \rightarrow 0$, the sample removed from the yoke (0") and the OCRI measured again ($B_{d,2}$). A flux reduction, $\Delta B_d = \frac{B_{d,1} - B_{d,2}}{B_{d,1}} \cdot (100)\ [\%]$, between about 0.4 and 1.2% was observed (see Table 24).
3) Sample was put back into the gap and subjected to two additional cycles $H_a = 0 \rightarrow -H_{d,av} \rightarrow 0$. It was removed after each cycle and OCRI measured. No significant additional loss was noted.

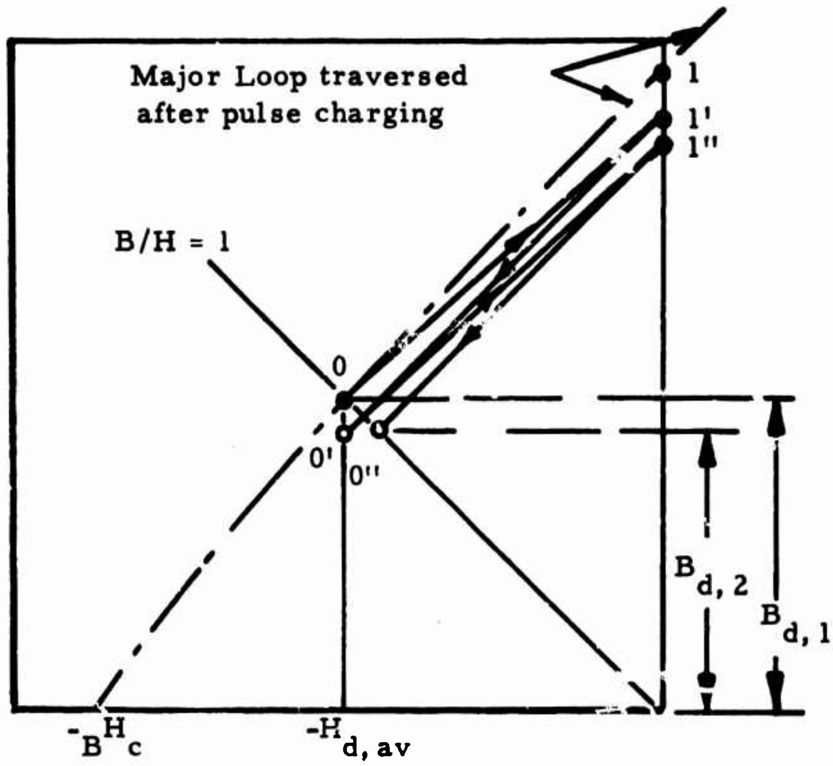

Figure 26a. Magnetic History of Sample During the First Slow Cycling Through Half Range.

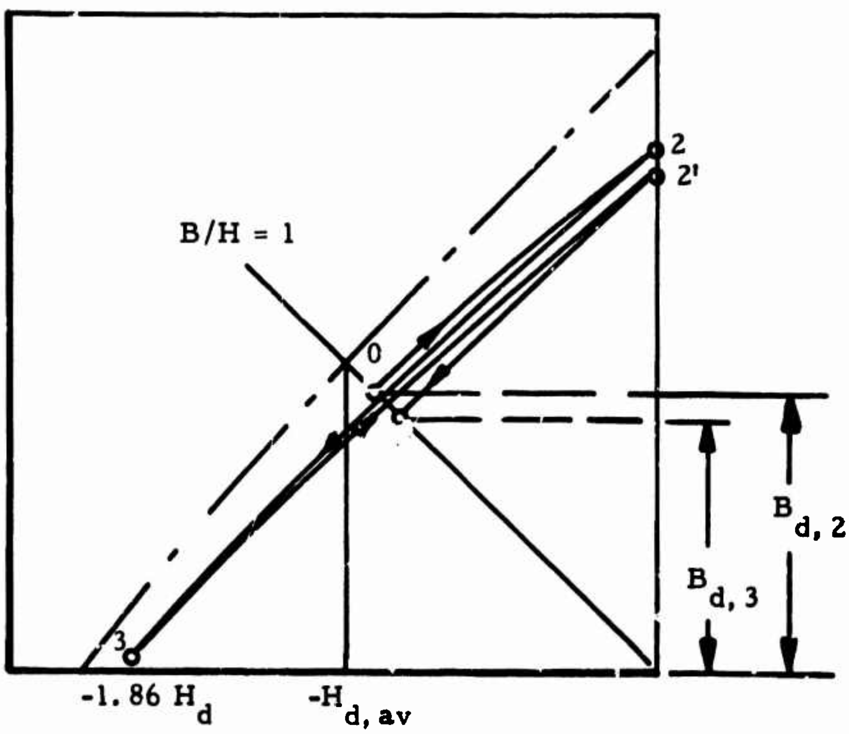

Figure 26b. Magnetic History of Sample During the First Slow Cycling Through Full Range.

B. Slow cycling through approximately the entire second quadrant. (See Figure 26b).

1) Initial OCRI value for this set of cycles is the final value of the 3 cycles of set A described above, and essentially the same as after the very first cycle ($B_{d,1}$). The new initial state 0 corresponds approximately to the old state 0'' of Figure 26a.

2) Sample was put into yoke, the field swept $H_a = 0 \rightarrow -1.86\ H_{d,av} \approx -0.93\ {}_BH_c \rightarrow 0$, the sample removed (0') and the OCRI value measured ($B_{d,3}$). An additional flux reduction was incurred, bringing the total "initial loss",

$$\delta B_d = \frac{B_{d,3} - B_{d,1}}{B_{d,1}} \cdot (100),$$ to between 2.4 and 4.7%.

3) Sample was put back into yoke and subjected to three additional full-swing cycles, $H_a = 0 \rightarrow -0.93\ {}_BH_c \rightarrow 0$. It was removed after each cycle and the OCRI measured. Again, no significant additional flux reduction was noted.

C. The samples were then put back into the yoke and the periodic cycling at a frequency of 1000 cph between the extremes of $-1.86\ H_{d,av} \approx -0.93\ {}_BH_c$ and $-0.14\ H_{d,av} \approx -0.07\ {}_BH_c$ began. This was carried to a total exposure of 2 million cycles, but cycling was interrupted at regular intervals - - after 20, 80, 1000 cycles, etc., - - the sample removed and the OCRI measured. The duration of these tests, counting "dead time" as well as cycling time, was approximately 2000 hours ("total elapsed time").

The losses for the 9 individual samples - - computed from the OCRI measurements after the first 3 half cycles, the following 4 slow full cycles, and the 2×10^6 full cycles at 1000 cph - - are presented in Table 24. Figure 27 shows the range of losses observed for the two sample types as a function of the number of cycles during the "a.c. exposure" phase, C. The flux values are normalized to the OCRI flux measured at the end of the first 4 full cycles traversed with manual field control. It can be seen that the isopressed samples had the lower losses and were very uniform within the group; the losses for the diepressed samples were about double those

TABLE 24

EFFECTS OF MAGNETIC FIELD CYCLING ON THE OPEN-CIRCUIT REMANENCE

AT $B/H \simeq 1$

Sample Number	Initial Losses *		Additional Loss **** After 2×10^6 cycles (%)	Initial Properties			
	**) (%)	***) (%)		$_M H_c$ (kOe)	H_k (kOe)	$_B H_c$ (Oe)	B_r (G)
A-82	0.59	2.36	0.16	43.0	8.50	7998	8824
A-86	0.42	2.63	0.16	45.5	9.02	7941	8721
A-91	0.36	2.57	0.16	44.6	8.56	7867	8614
A-99	0.47	3.75	0.16	54.7	7.45	7905	3836
B-75	0.71	3.37	0.31	35.6	6.07	6877	7775
B-86	1.21	4.65	0.32	37.5	6.79	6791	7620
B-94	0.77	3.61	0.25	32.9	7.26	6887	7590
B-96	0.59	4.08	0.37	30.4	6.26	6861	7803

*) Expressed in % of initial value of the OCRI

**) Measured after 3 cycles $H = 0 \rightarrow -H_d \rightarrow 0$

***) Measured after 4 additional cycles $H = 0 \rightarrow -1.86\ H_d \rightarrow 0$

****) Frequency 1000 cph, amplitude $\pm$ 0.86 H_d over bias of $-H_d$. Loss in % of value measured after the first 7 slow cycles.

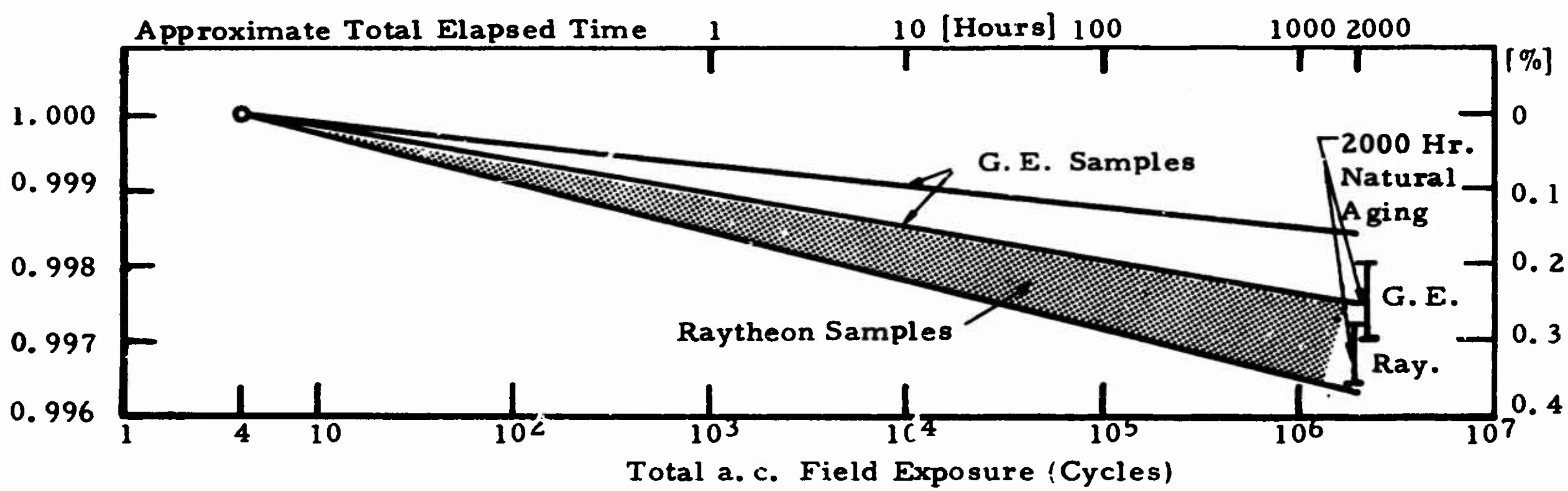

Figure 27. Long-Term Losses During Cyclic Magnetic Field Test.

of the isopressed ones and they covered a wider band. In either case, the OCRI reduction after a given total elapsed time period is about the same as it was in the previously described "natural air aging" experiments done without an alternating field. To facilitate comparison, we have added the approximate elapsed time scale on the top of Figure 27 and indicated the range of loss observed after 2000 hours of natural air aging of non-prestabilized magnets with two bars on the right side of the graph.

The obvious conclusion is that exposure of good, square-loop $SmCo_5$ sintered magnets to large alternating magnetic fields has no effect on the long-term stability of the operating flux, even if the negative peak field is as high as ${}_BH_c$ and thus B is reduced to zero. The final, stable recoil loop is established the first time the demagnetizing field reaches its maximum value (at least if several seconds are allowed for this first negative half-cycle, as in our experiments). Even on the second and third cycles, the magnet operates reversibly on the same recoil loop within our limit of flux resolution, about 0.05%.

Only when a previous negative field peak is later exceeded, a measurable additional drop of the OCRI occurs.